# Nuclear Energy Research Initiative

## 2002 Annual Report

Available to DOE, DOE contractors, and the public from the

U.S. Department of Energy
Office of Nuclear Energy, Science and Technology
1000 Independence Avenue, S.W.
Washington, D.C. 20585

# Table of Contents

# Foreword

The Nuclear Energy Research Initiative (NERI) began in Fiscal Year (FY) 1999 as the core of a new, restructured Federal effort to develop advanced nuclear energy concepts and technologies. NERI grew out of the recommendations made in the November 1997 report by the President's Committee of Advisors on Science and Technology on Federal energy research and development (R&D). FY 2002 marks the completion of the first round of NERI sponsored research projects initiated in FY 1999. The results and accomplishments of these 46 initial projects is a major focus of this year's NERI Annual Report.

Since its inception the NERI program has focused on innovation and forward looking technological advances. NERI has also gained momentum in addressing issues associated with the maintenance of existing U.S. nuclear generating plants, as well as with other areas identified in the National Energy Policy. In combination with the Nuclear Power 2010 and Generation IV Nuclear Energy Systems initiatives, NERI will respond to the Nation's need for new electricity generating capacity in an economical and environmentally friendly manner.

NERI has been realizing its goals to develop advanced nuclear energy systems, and to provide state-of-the-art information concerning nuclear technology and science. The research effort conducted by the Nation's universities, laboratories, and industry partners has helped to maintain the nuclear research infrastructure in this country and has focused attention on the United States as a nuclear R&D leader. The NERI program continues to use independent, expert, peer reviewers to competitively select project proposals from a wide range of researchers.

This Annual Report summarizes research progress based on information submitted by the principal investigators for NERI projects initiated in FY 1999, FY 2000, and FY 2001. Also included in this document are the abstracts for the FY 2002 NERI research awards. This report disseminates the results of NERI-sponsored research to the wide R&D community to spur yet more innovation, assuring a bright future for nuclear energy in the United States and the world.

William D. Magwood IV, Director
Office of Nuclear Energy, Science and Technology
U.S. Department of Energy

NUCLEAR ENERGY RESEARCH INITIATIVE

# I. Introduction

The United States Department of Energy (DOE) created the Nuclear Energy Research Initiative (NERI) in 1998 in response to recommendations provided by the President's Committee of Advisors on Science and Technology (PCAST). The purpose of NERI is to sponsor research and development (R&D) in the nuclear energy sciences to address the principal barriers to the future use of nuclear energy in the United States. NERI will also help preserve the nuclear science and engineering infrastructure within the Nation's universities, laboratories, and industry, and will advance the development of nuclear energy technology, enabling the United States to maintain a competitive position worldwide. It is the position of DOE that funding research at the Nation's research institutions and corporations will create solutions to important nuclear issues and that a new potential for nuclear energy will emerge in the United States. The DOE Office of Nuclear Energy, Science and Technology (NE) funds and administers the NERI program.

The Nuclear Energy Research Initiative Annual Report serves to inform interested parties of progress made in NERI on a programmatic level as well as research progress made in individual NERI projects. Section 2 of this report provides more background on the creation and implementation of NERI, on the NERI mission and its goals and objectives, on the focus areas for NERI research, and on the management of the program. Section 3 provides some brief summaries of NERI research efforts and highlights the major accomplishments of the NERI program.

Sections 4 through 7 provide summary project status reports by research focus area. These status reports discuss NERI projects that have advanced sufficiently to enable DOE to report on their progress. Research objectives, progress made over the last two years since the initial report, and activities planned for the next year are described for each project. Also included in these chapters are the corresponding abstracts for the latest FY 2002 NERI research awards. Project numbers are designated by the fiscal year (FY) in which their proposal is submitted and the award was made. However, the bulk of the first year's research effort is typically completed in the subsequent fiscal year. At the end of the document there is an Index of NERI Projects sequentially ordered by FY and project number.

NUCLEAR ENERGY RESEARCH INITIATIVE

## 2. Background

In January 1997, the President tasked his Committee of Advisors on Science and Technology (PCAST) to review the current national energy R&D portfolio and to provide a strategy to ensure that the United States has a program to address the Nation's energy and environmental needs for the next century.

In its November 1997 Report to the President, Federal Energy Research and Development for the Challenges of the Twenty-First Century, the PCAST panel on Energy Research and Development determined that it was important to establish nuclear energy as a viable and expandable option and that a properly focused R&D effort to address the potential long-term barriers to expanded use of nuclear power (e.g., nuclear waste, proliferation, safety and economics) was appropriate. The PCAST panel further recommended that DOE reinvigorate its nuclear energy R&D activities in order to address these potential barriers with a new nuclear energy research initiative. DOE would fund research through this new initiative, based on competitive selection of proposals from the national laboratories, universities, and industry.

DOE endorsed the PCAST recommendations and received Congressional appropriations in FY 1999, allowing NERI to sponsor innovative scientific and engineering R&D to address the key issues affecting the future use of nuclear energy and preserve the Nation's nuclear science and technology leadership.

In 1999, the PCAST report, The Federal Role in International Cooperation on Energy Innovation, recommended creation of an international component to NERI to promote "bilateral and multilateral research focused on advanced technologies for improving the cost, safety, waste management, and proliferation-resistance of nuclear fission energy systems." In FY 2001, the Department launched the new International Nuclear Energy Research Initiative (I-NERI), for bilateral and multilateral nuclear energy research. Approximately $15 million has been appropriated for bilateral, cost-shared research work under the I-NERI program with South Korea and France. A third collaboration involves Argonne National Laboratory and a consortium of ten international participants represented by the U.S. Nuclear Regulatory Commission (NRC) and the European Organization for Economic Co-operation and Development (OECD) with offices throughout the world. Similar international agreements with other countries are being considered. I-NERI allows DOE to leverage federal investment with international resources through cost-share arrangements with each participating country on a wide range of nuclear technology topics. I-NERI will further enhance the influence of the United States and DOE in international policy discussions on the future direction of nuclear energy. Similar to NERI, I-NERI features competitive, researcher-initiated R&D selected through an independent peer-review process by international experts from the United States and its partners. A separate report covering the research effort provided by I-NERI is expected to be published in early 2003.

### NERI Development

In order to determine the initial focus of the NERI research areas, DOE convened a workshop of nuclear community stakeholders in April 1998, representing national laboratories, universities, and industry. As a result of this NERI workshop[1], DOE focused its initial scientific and engineering R&D in the following areas:

- Proliferation-resistant reactors and fuel technology
- New reactor designs to achieve improved performance, higher efficiency, and reduced cost, including low-output power reactors for use where large reactors are not attractive
- Advanced nuclear fuels
- New technologies for management of nuclear waste
- Fundamental nuclear science

To encourage innovative R&D, a unique process for selecting new NERI projects has been employed since the program's inception. In response to the NERI solicitations,

[1]Summary Report of the Nuclear Energy Research Initiative Workshop-An Assessment of Research Opportunities for Nuclear Energy, Technology and Education, June 1998.

principal investigators (PIs) select research topics of interest and define the scope and extent of the R&D in their proposals. DOE employs an independent, expert peer review process to judge the scientific and technical merit of the R&D proposals. For those proposals judged to have the highest scientific and technical merit, DOE conducts a programmatic review to ensure conformance of selected projects with DOE policy and programmatic requirements. The two reviews result in award-selection recommendations to DOE's Source Selection Official.

In the intervening years since the initiation of NERI a number of influencing events have occurred which have helped shape and focus NERI research activities.

- In 1998, DOE established the independent Nuclear Energy Research Advisory Committee (NERAC) to provide advice to the Secretary and to the Director, Office of Nuclear Energy, Science, and Technology (NE) on the DOE civilian nuclear technology program. In June 2000, NERAC issued the Long Term Nuclear Technology Research and Development Plan, which identifies the research and technology development necessary over the next 10-20 years to help ensure the long-term viability of nuclear energy as an electricity generation option. NERAC also established a task force to identify R&D needs related to nuclear non-proliferation issues associated with nuclear power production. Their recommendations for appropriate research in this area were provided to DOE in a January 2001 report titled, Technical Opportunities to Increase the Proliferation Resistance of Global Civilian Nuclear Power Systems (TOPS).
- The National Energy Policy, issued in May 2001 by the Vice President's National Energy Policy Development Group, supports the expansion of nuclear energy as one of its major initiatives for meeting the growing energy requirements of the United States. The National Energy Policy provides the core element in the planning for DOE's nuclear energy research programs addressing, among other areas, the research and development of advanced reactor and fuel cycle concepts, hydrogen production from nuclear energy, and the associated enabling sciences and technologies.
- In September 2002, NERAC issued the Draft Technology Roadmap for Generation IV Nuclear Energy Systems. In coordination with the ten-member country Generation IV International Forum (GIF) six reactor system concepts were selected for further research and development. These include the Very-High-Temperature Reactor System, the Gas-Cooled Fast Reactor System, the Supercritical Water-Cooled Reactor System, the Lead-Cooled Fast Reactor System, the Sodium-Cooled Fast Reactor System, and the Molten Salt Reactor System.

**NERI Mission**

The importance of nuclear power to the World's future energy supply requires that DOE apply its unique resources, specialized expertise, and national leadership to address key issues affecting the future of nuclear energy. NERI is a national research-oriented initiative focused on innovation and competitiveness that brings together national laboratories, universities, and industry to explore and develop new nuclear power technology. In so doing, NERI advances the state of scientific knowledge and promotes an enhanced domestic nuclear energy research and science infrastructure at universities, national laboratories, and industry that directly supports DOE's energy mission-a Secretarial priority.

The NERI program also supports the National Energy Policy by conducting research to advance the state of nuclear science and technology in the United States. This research addresses the key technical issues impacting the expanded use of nuclear energy. NERI is essential to helping DOE foster innovative ideas in such areas as advanced nuclear energy systems, hydrogen production from nuclear power, advanced nuclear fuels and fuel cycles, and fundamental science. This research enhances the ability of nuclear energy to help meet the Nation's future energy needs and environmental goals. To achieve these long-range goals, NERI has the following objectives:

- Address and help overcome the potential technical and scientific obstacles to the long-term future use of nuclear energy in the United States, including those involving nonproliferation, economics, and nuclear waste disposition.
- Advance the state of U.S. nuclear technology so that it can maintain a competitive position in overseas markets and a future domestic market.
- Promote and maintain a nuclear science and engineering infrastructure to meet future technical challenges.

Working in tandem with the Nuclear Power 2010 program[2], the Generation IV Nuclear Energy Systems Initiative[3], and I-NERI, the NERI program supports the following NE programmatic goals:

- Facilitate an additional 50,000 megawatts (MW) of electricity by 2020, saving the consumer $500 million annually in comparison to producing the same electricity using fossil fuel.
- Avoid the emissions of 82 million metric tons of carbon, 2.5 million tons of sulfur dioxide, and 1.2 million tons of nitrogen oxide into the Earth's atmosphere each year.

This latter activity will make a significant contribution towards achieving the President's Global Climate Change goal of reducing greenhouse gas intensity by 18 percent by 2012.

**NERI Work Scope**

In FY 2002, NERI expanded its R&D focus to address new research requirements introduced in the National Energy Policy. The following paragraphs defined the NERI research areas:

<u>Advanced Nuclear Energy Systems:</u> This program element includes the investigation and preliminary development of advanced concepts for reactor and power conversion systems. These systems offer the prospect of improved performance and operation, design simplification, enhanced safety, and reduced overall cost. Projects involve innovative reactors, system and component designs, alternative power conversion cycles for terrestrial applications, new research in advanced digital instrumentation and control and automation technologies, and other important design features and characteristics.

<u>Hydrogen Production from Nuclear Power</u>: This program element includes research and development to identify and evaluate new and innovative concepts for producing hydrogen using nuclear reactors. This research includes investigation of hydrogen generation processes compatible with advanced reactor systems, and the integrating parameters needed to develop systems that are efficient and cost-effective overall.

<u>Advanced Nuclear Fuels/Fuel Cycles</u>: This element includes research and development to provide measurable improvements in the understanding and performance of nuclear fuel and fuel cycles with respect to safety, waste production, proliferation resistance, and economics, to enhance the long-term viability of nuclear energy systems. This research includes improvements in the performance of fuels for advanced systems, and development of fuels capable of withstanding the conditions in the supercritical light water reactor (LWR) regime and of advanced proliferation resistant-fuels capable of high burn-up such as those needed in support of the Generation IV concepts.

<u>Fundamental Science</u>: This element includes research and development in the fields of materials science and fundamental chemistry. Fundamental science research funded by NERI applies to and supports one or more of the preceding program elements in advanced nuclear engineering technology. Material sciences applications include research and development on materials for use in advanced nuclear reactor systems, structures, and components, including fuel cladding that may perform in high-radiation fields, high-temperatures and pressures, and/or in highly corrosive environments (i.e., lead-bismuth). Chemical science research may focus on development and improvement of primary and secondary coolant chemistry in advanced reactors. Other research subjects include the investigation of nuclear isomers that could prove beneficial in civilian applications.

In general, it should be noted that safety, nonproliferation, and waste management are considerations intrinsic to the above research topics, especially for the advanced nuclear energy systems and advanced fuels/fuel cycles. Thus, they become selection criteria across all four focus areas, and do not in themselves constitute focus areas.

The full-page graphic on the following page summarizes the key features of the NERI program.

[2] The Nuclear Power 2010 program was implemented in FY 2002 to address nuclear regulatory, technical, and institutional issues to enable one or more orders to be placed for new commercial nuclear power plants in the United States by 2005 for deployment by 2010.

[3] The Generation IV Nuclear Energy Systems Initiative was implemented in FY 2000 to focus on the development and demonstration of one or more Generation IV nuclear energy systems that offer advantages in the areas of economics, safety and reliability, and sustainability, and that could be deployed commercially by 2030.

# An Overview of the NERI Program

## Vision

To maintain a viable nuclear energy option to help meet the Nation's future energy needs

## Goals/Objectives

- Address potential technical and scientific obstacles
- Advance the state of U.S. nuclear technology
- Promote and maintain a nuclear infrastructure

## Scope of Program

- Advanced Nuclear Energy Systems
- Advanced Nuclear Fuels/Fuel Cycles
- Hydrogen Production from Nuclear Power
- Fundamental Science

## R&D Priorities

- Innovative reactors, systems, and components
- Hydrogen generation
- Alternative power conversion cycles
- High-burnup, proliferation-resistant fuels
- Advanced digital instrumentation & controls
- Supercritical Light Water Reactor fuels
- Automation technologies
- Corrosion-resistant fuel cladding
- Advanced structural materials
- Enhanced coolant chemistry

## Implementation Strategy

- Competitive peer-reviewed R&D selection process
- Individually managed projects
- Collaborative research efforts
- Continuous DOE oversight

## Results

- U.S. nuclear leadership
- Innovative technologies
- Nuclear infrastructure development
- Advances in fundamental science
- Nuclear public awareness
- Worldwide partnerships

NUCLEAR ENERGY RESEARCH INITIATIVE

## 3. NERI Accomplishments

This section discusses the program's progress in attracting research proposals, awarding annual R&D funding, facilitating the successful completion of the initial NERI-funded projects, and increasing the number of students participating in nuclear-related studies and research.

### Project Awards

In FY 1999, DOE's NERI program received 308 R&D proposals from U.S. universities, national laboratories, and industry in response to its first solicitation. The initial FY 1999 procurement was completed with the awarding and issuing of grants, cooperative agreements, and laboratory work authorizations for 46 R&D projects. The proposed research represented participants from 45 institutions and organizations. Thirty-two of the projects involved collaborations of multiple organizations. Eleven foreign R&D organizations also participated in NERI collaborative projects. The duration of these annually funded projects was one to three years, with the majority lasting three years. The total cost of these 46 research projects for the three-year period was approximately $52 million.

Figure 1 depicts the number of research projects in each of the four R&D areas awarded in FY 1999. Proliferation-resistant technologies, though not considered separately, are incorporated in most of the research projects on advanced nuclear fuels and new reactor designs and technologies. In addition, the fundamental nuclear science area includes research projects in materials science, fundamental chemistry, computational and engineering science, and nuclear physics.

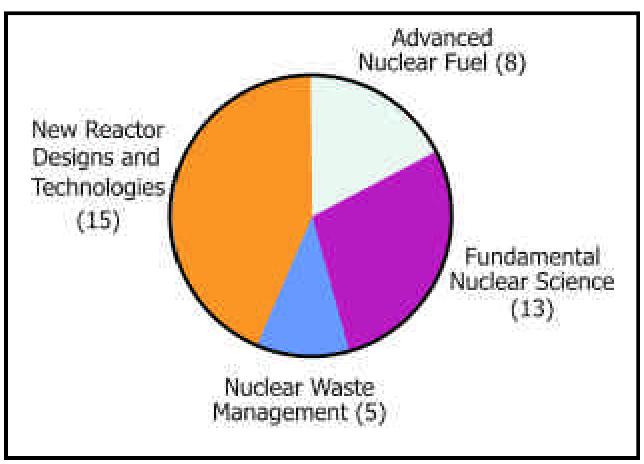



Figure 1. The graph illustrates FY 1999 NERI projects by R&D areas.

In FY 2000, FY 2001, and FY 2002, scientific and technical development advanced through the continuation of research efforts begun in FY 1999 as well as through the initiation of new awards.

- In FY 2000, 10 NERI R&D projects were awarded involving 18 U.S. and 6 foreign R&D organizations.
- In FY 2001, 13 NERI R&D projects were awarded involving 23 U.S. and 5 foreign R&D organizations.
- In FY 2002, 24 projects were awarded involving 32 U.S. and 5 foreign R&D organizations.

Figure 2 illustrates the cumulative total of research projects for FY 2000, FY 2001, and FY 2002 in each of the three major R&D areas. Nuclear Waste Management was discontinued as a focus research area for NERI after the FY 1999 award cycle.

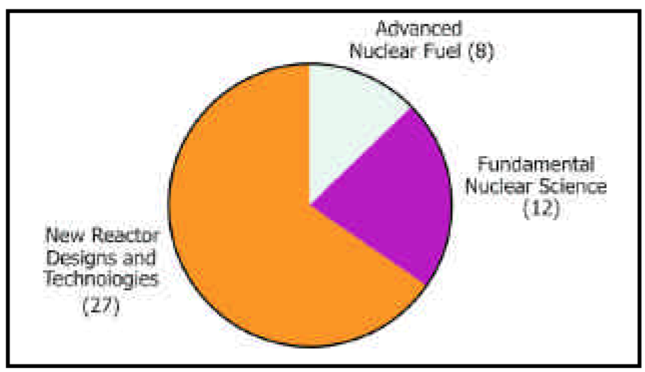



Figure 2. The graph indicates the division of NERI projects by R&D area for FY 2000, FY 2001, and FY 2002.

Funding for NERI is appropriated annually by Congress in the Energy and Water Development Appropriations Act.

- NERI funding for FY 1999 was a total of $19 million with $17.5 million available for new awards.
- Funding for FY 2000 was $21.5 million, which provided for Year 2 funding of FY 1999 awards in addition to approximately $2.7 million for new FY 2000 awards.

- FY 2001 funding was $26.5 million, which provided for Year 3 funding of FY 1999 awards, Year 2 funding for FY 2000 awards, and approximately $5.7 million for 13 new FY 2001 awards.
- In FY 2002, NERI project funding was $25.6 million with approximately $10 million allocated for the new awards and $9.3 million for continuing ongoing research projects begun in FY 2000 and FY 2001.

To date, over $110 million has been awarded to fund NERI research projects. Figure 3 shows the distribution of these funds among the national laboratories, U.S. universities, and industry.

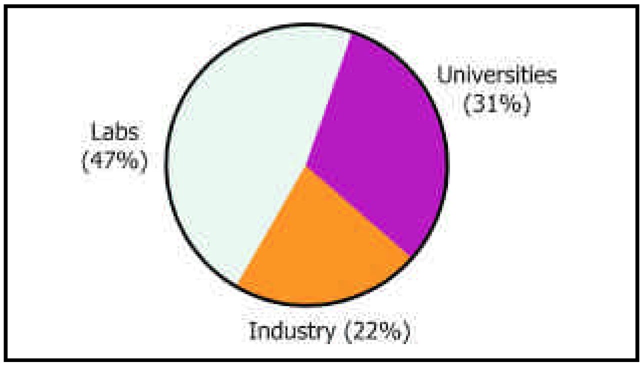



Figure 3. The chart illustrates the overall distribution of NERI funds for FY 1999 through FY 2002.

DOE has not funded foreign participants in existing projects as part of the NERI program. Rather, their participation has been supported by the foreign organizations interested in the research being conducted. Although the principal investigators have been responsible for soliciting such support, foreign participation in NERI projects is contingent upon DOE approval.

### FY 1999 Project Completions

This year marked the scheduled completion of the initial 46 FY 1999 NERI projects. Based on reported accomplishments, it is clear that NERI's stated goals and objectives are being met. These collaborative efforts between the public and private sectors have resulted in significant enhancements in the U.S. nuclear science and engineering infrastructure, especially in the areas of human and physical resources and capabilities. This will allow the United States to better meet future technical challenges related to nuclear energy. NERI research includes collaboration with over 25 international organizations. These efforts, coupled with those of I-NERI and the Generation IV Nuclear Systems Initiative, have served to revive the Nation's leadership role in international nuclear R&D. Moreover, the technology advances will allow the United States to maintain a competitive position in overseas energy markets and a future domestic market.

Finally, through accomplishment of their stated research objectives, these NERI projects have addressed and helped to overcome a number of potential technical and scientific obstacles to the long-term future use of nuclear energy in the United States. A few examples of the FY 1999 project accomplishments in each area of emphasis are described below. Additional details on individual project accomplishments are contained in the project summaries located in the following chapters.

Designing a Low-Cost, Proliferation-Resistant Reactor

- Westinghouse led an international team that is developing an innovative proliferation-resistant, water-cooled reactor called IRIS (International Reactor Innovative and Secure). IRIS is a new modular, passively safe, natural circulation (i.e., with no active pumps) design that is based on proven light water reactor experience. Detailed reactor physics and heat transfer analyses have been performed. Preliminary fuel and materials selections have been made for the initial conceptual design. Component manufacturers, an architectural engineering firm, and engineering analysis teams have begun further development of a detailed design that enhances passive safety and proliferation-resistance and that can be built at a reduced cost. (Project No. 99-027)

Automating Future Nuclear Power Plants

- Oak Ridge National Laboratory (ORNL) led a team that is developing new advanced controls, diagnostic techniques, and information systems that could be used to automate future nuclear plants. The team is developing, testing, and implementing "adaptive" control strategies that recognize changes in operating data and use a decision-making module to detect system faults. (Project No. 99-119)
- Pacific Northwest National Laboratory (PNNL) focused on an on-line, intelligent, self-diagnostic monitoring system for next-generation nuclear plants, using wireless radio-frequency (RF) tagging technology. The wireless RF sensors, communication modules, and hardware and software allow RF sensors to be placed inside or near individual plant system components to provide fault detection and condition-monitoring information. (Project No. 99-168)

Bringing Advanced Ceramic Materials to Nuclear Power Applications

- The University of Florida has developed a silicon carbide (SiC) radiation-resistant material with excellent thermodynamic properties that will improve the economics of nuclear fuel. A SiC coating was applied to fuel cladding and several experiments have been completed that show how this SiC coating behaves in high-temperature environments. (Project No. 99-229)
- PNNL has explored how various ceramics behave in different thermal environments to determine whether SiC fibers, monolithic materials, and composite materials could be used in very high-temperature fission reactors under harsh irradiation environments. (Project No. 99-281)
- Gamma Engineering led a team to investigate the use of a continuous fiber ceramic composite cladding for commercial water reactor fuel. Researchers developed, tested, and irradiated the ceramic cladding material and demonstrated its ability to handle harsh accident conditions. This completed NERI project has served to demonstrate that new cladding concepts are possible. This new ceramic composite was further developed as part of a DOE-sponsored Phase I Small Business Innovation Research (SBIR) commercialization effort. (Project No. 99-224)

Developing New, Proliferation-Resistant Nuclear Fuels

- The Idaho National Engineering and Environmental Laboratory and eight research organizations investigated the feasibility of using thorium/uranium (Th/U) dioxide ceramic fuels to increase fuel utilization and proliferation-resistance in light water reactors. The team is developing Th/U core designs, fuel pellet fabrication methods, and temperature-dependent Th/U fuel property correlations, and is determining the corrosion and oxidation rate characteristics of the Th/U waste form. (Project No. 99-153)
- The Argonne National Laboratory (ANL) and Purdue University investigated the use of metallic Th/U fuel that has better thermal and proliferation-resistant properties and that can be disposed of directly. Initial physics and heat transfer calculations, laboratory experiments, and production of different Th/U fuel mixtures have been completed. Further research is focusing on the possible use of Th/U mixtures in new fuel designs in order to reduce the relative amount of plutonium isotopes produced in power reactors. (Project No. 99-095)

Developing Radiation-Resistant Alloys

- PNNL worked with General Electric (GE) and the University of Michigan to develop new alloys that are resistant to radiation damage. By doping common stainless steels with small amounts of platinum, hafnium, and other elements, it may be possible to reduce radiation-induced damage mechanisms, such as stress corrosion-cracking, which limit reactor component lifetimes. The team has irradiated materials with nickel ions and protons to simulate accelerated irradiation damage in reactor metals. Tests have been completed that show how these doped metals perform at different temperature and chemistry conditions. (Project No. 99-280)

Exploring Direct Energy Conversion Technologies for Nuclear Power

- Sandia National Laboratories (SNL) led a team investigating direct fission energy conversion concepts using reactor pumped lasers, pulsed power, space technology, solid state converters, magnetohydrodynamics, and direct radioactive isotope decay technologies. Three promising concepts have been selected for further development. Additional physics analysis and testing have been initiated. Critical technology R&D needs have been identified for future test programs, and for the Generation IV Technology Roadmap R&D effort. (Project No. 99-199)
- ORNL worked on developing an advanced reactor concept that uses an ORNL-developed graphite foam material with superior heat transfer characteristics. Irradiation tests at ORNL's High Flux Reactor have demonstrated that this graphite foam can withstand extreme neutron damage and still provide superior heat transfer capability. A simple nuclear "battery" reactor with nuclear fuel dispersed within the light graphite foam would allow for new and remote applications. (Project No. 99-064)

Reactor Physics Experiments for Advanced Nuclear Power Systems

- Experimental measurements of reactor physics data for lead-cooled fast reactors and nuclear waste transmutation systems have being conducted by ANL in collaboration with the French Atomic Energy Commission (CEA) at the Caderache facilities. The Argonne team has completed the development, planning, and implementation of no-cost experiments in France that will lead to valuable physics information needed for advanced Generation IV reactor designs. The experiments are being used for formal reactor physics calculation benchmarks to validate U.S. and international reactor engineering computer codes and cross section data bases. (Project No. 99-039)

- Two experimental projects at SNL will provide important information on criticality safety and spent fuel shipment safety, needed for advanced reactor fuel designs that use higher initial fuel enrichments and have larger end-of-life fuel burn-up exposures than currently-operating plants. The data generated from these experiments will serve to verify computational methods used for reactor safety calculations, reduce the licensing burden on future reactor designs, and remove overly conservative assumptions made in the absence of such data. (Project No. 99-200)

## U.S. University Involvement

Twenty-eight U.S. universities participate either as lead investigators or collaborators in about 75 percent of the 93 NERI projects that have been funded. Figure 3 indicates the location and names of the participating institutions. A significant increased enrollment of students across the Nation in nuclear-related fields is attributed to the research opportunities provided by NERI at these universities.

University student participation has been at all levels- in undergraduate, master's, and doctoral degree programs. Figure 4 is a summary of student participation by degree

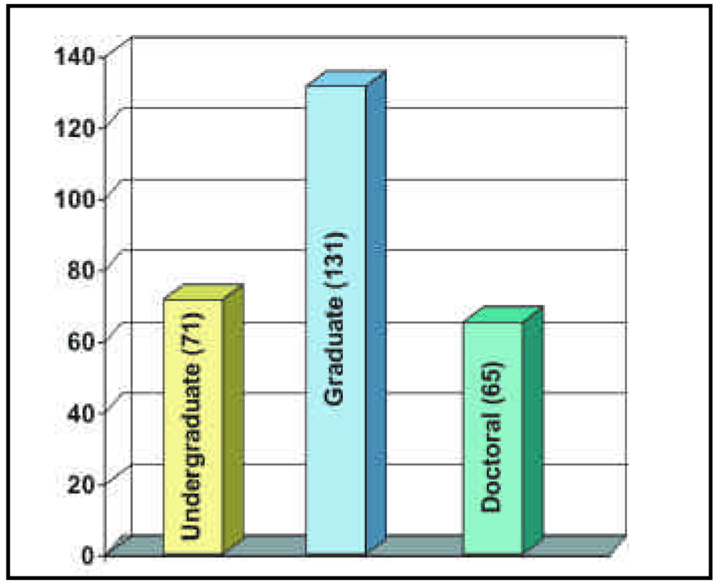
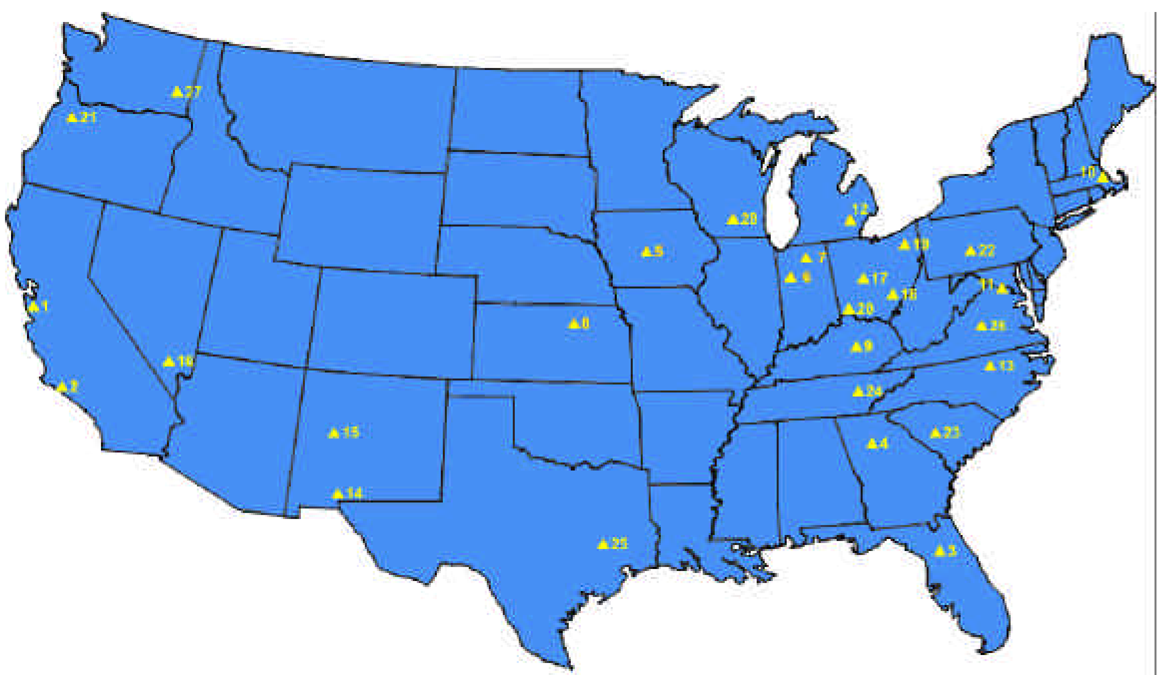



Figure 4. The graph indicates the number of university students participating in NERI projects in each type of academic degree program.



**UNIVERSITY KEY**

1. University of California, Berkeley
2. University of California, LA
3. University of Florida
4. Georgia Institute of Technology
5. Iowa State University
6. Purdue University
7. University of Notre Dame
8. Kansas State University
9. University of Kentucky
10. Massachusetts Institute of Technology
11. University of Maryland, College Park
12. University of Michigan
13. North Carolina State University
14. New Mexico State University
15. University of New Mexico
16. University of Nevada, LV
17. Ohio State University
18. Ohio University
19. University of Akron
20. University of Cincinnati
21. Oregon State University
22. Penn. State University
23. University of South Carolina
24. University of Tennessee
25. Texas A&M University
26. University of Virginia
27. Washington State University
28. University of Wisconsin

Figure 3. The map shows the state locations for universities participating in NERI projects; the key names the universities.

program at individual universities associated with the FY 1999, 2000, and 2001 NERI projects. A total of 267 students have participated in 49 of the 69 projects funded during these three years. In addition, numerous post-doctoral participants have been involved at these universities in NERI research projects. The number of students participating in the newly awarded FY 2002 projects is yet to be determined.

### Other NERI Participants

In addition to the 28 U.S. universities noted in the previous section, since FY 1999 NERI research participants have included 11 national laboratories, 2 government agencies, 27 private businesses, and 28 foreign organizations. The names of these participating organizations are provided in the following tables.

**U. S. Department of Energy Laboratories**
Ames Laboratory
Argonne National Laboratory
Brookhaven National Laboratory
Idaho National Engineering and Environmental National Laboratory
Lawrence Berkeley National Laboratory
Lawrence Livermore National Laboratory
Los Alamos National Laboratory
Oak Ridge National Laboratory
Pacific Northwest National Laboratory
Sandia National Laboratory
Savannah River Technology Center

**Government Agencies**
National Institute of Standards and Technology
U.S. Nuclear Regulatory Commission

**Industrial Organizations**
Bechtel
CEGA Corporation
Dominion Generation
Duke Engineering
Egan Associates
Electric Power Research Institute
Entergy Nuclear, Inc.
Florida Power and Light
Framatome ANP, Inc.
continued

**Industrial Organizations (continued)**
Gamma Engineering
General Electric
General Electric Global Research Center
General Atomics
Global Nuclear Fuel
McDermott Technologies
Newport News Ship Building and Drydock Co.
Northern Engineering and Research
Pacific Sierra
Pacific Southern Electric and Gas
Panlyon Technologies
Rockwell Science Center
Siemens Power Corporation
SRI International
Swales Aerospace
Tennessee Valley Authority
Westinghouse Electric Company
(n,p) Energy, Inc.

**International Collaborators**
Atomic Energy of Canada (Canada)
Ben Gurion University (Israel)
British Nuclear Fuel (UK)
Chosun University (Korea)
Commissariat a l'Energie Atomique (France)
Framatome (France)
Forschungszentrum (Germany)
Hitachi (Japan)
Imperial College of London (United Kingdom)
Institute of Physics and Power Engineering (Russia)
Italian National Agency for New Technologies, Energy and Environment (ENEA)
Japan Nuclear Cycle Development Institute (Japan)
Japan Atomic Power Company (Japan)
Kurchatov Institute (Russia)
Mitsubishi Heavy Industries (Japan)
National Atomic Energy Commission and University of Cuyo (Argentina)
OECD Nuclear Energy Agency (France)
PBMR, Ltd. (South Africa)
Polytechnical Institute of Milan (Italy)
Studsvik Scanpower Inc. (Sweden)
Tokai University (Japan)
Tokyo Institute of Technology (Japan)
Toshiba (Japan)
Toyama University (Japan)
University of Manchester (UK )
University of Rome (Italy)
University of Tokyo (Japan)
VTT Manufacturing Technology (Finland)

## 4. Advanced Nuclear Energy Systems

This program element includes 47 research projects to date of which 20 were awarded in FY 1999, 8 in FY 2000, 7 in FY 2001, and 12 in FY 2002. It includes the investigation and preliminary development of advanced concepts for reactor and power conversion systems. These systems offer the prospect of improved performance and operation, design simplification, enhanced safety, and reduced overall cost. Projects may involve innovative reactors; system or component designs; alternative power conversion cycles for terrestrial applications; new research in advanced digital instrumentation and control and automation technologies; hydrogen production from nuclear reactors; and identification and evaluation of alternative methods, analyses, and technologies to reduce the costs of constructing future nuclear power plants.

Additionally, this element includes research projects to improve the intrinsic proliferation-resistant qualities of advanced reactors and fuel systems. Possible technology opportunities and subjects of investigation include alternative proliferation-resistant reactor concepts, systems that minimize the generation of weapons-usable nuclear materials (e.g., Pu-239) and waste by-products, or systems that increase energy extraction from the utilization of plutonium and other actinide isotopes generated in the fuel.

Projects involving advanced reactors under this program element specifically address, among other items, the characteristics, feasibility, safety features, proliferation-resistance, and economic competitiveness of reactor systems, and additional research that may be required. These reactor concepts include advancements in light water reactor technology to achieve higher performance, or development of other higher temperature advanced reactor designs for higher efficiencies.

Other advanced-reactor concepts include compact or modular reactor designs suitable for transport to remote locations, and alternative energy production or co-generation reactor applications. Desirable features include long-lived reactor cores that minimize or avoid altogether the need for refueling, and concepts that maximize fuel burn-up or employ advanced energy conversion technology.

Finally, this program element includes research and development to identify and evaluate new and innovative concepts for producing hydrogen using nuclear reactors. This research includes investigation of hydrogen generation processes compatible with advanced reactor systems, and the integrating parameters needed to develop systems that are efficient and cost-effective overall.

# Nuclear Energy Research Initiative

Directory Advanced Nuclear Energy Systems Project Summaries and Abstracts

NUCLEAR ENERGY RESEARCH INITIATIVE

# Application of Innovative Experimental and Numerical Techniques for the Assessment of Reactor Pressure Vessel Structural Integrity

Primary Investigator: T.Y. Chu, Sandia National Laboratories (SNL)

Collaborators: U.S. Nuclear Regulatory Commission (NRC); Organization for Economic Cooperation and Development (OECD)/Nuclear Energy Agency (NEA)

Project Number: 99-018

Project Start Date: August 1999

Project End Date: March 2003

## Research Objectives

The Nuclear Energy Research Initiative (NERI)/NRC/OECD sponsored program consists of eight international partners: Belgium, Czech Republic, Finland, France, Germany, Spain, Sweden, and the United States. U.S. support is provided by the NRC and the Department of Energy's NERI program. This experimental/analytical program builds on the accomplishments of a previous NRC-sponsored Lower Head Failure (LHF) program (NUREG/CR-5582). The current program is referred to as the OECD Lower Head Failure (OLHF) program to distinguish it from the previous program and to recognize the international participation of the OECD.

This project consists of both experimental and analytical efforts in investigating the structural integrity of reactor pressure vessels. Experiments simulating the thermal/mechanical loads to a reactor pressure vessel generate data that can be implemented into a finite element code, such as the commercially available code ABAQUS, to assess the ability of the code to capture the response of the pressure vessel to severe accident conditions. In addition, the pressure vessel material (SA533B1 steel) used in these experiments is prototypical of reactor pressurized water reactor vessel material and is well-characterized by material property testing as part of this program.

The lower head of the reactor pressure vessel (RPV) can be subjected to significant thermal and pressure loads in the event of a core meltdown accident. The mechanical behavior of the reactor vessel's lower head is of importance both in severe accident assessment and the assessment of accident mitigation strategies. For severe-accident assessment, the failure of the lower head defines the initial conditions for all ex-vessel events, and in accident mitigation the knowledge of mechanical behavior of the reactor vessel defines the possible operational envelope for accident mitigation. The need for validated models of the lower head in accident scenarios is accomplished by well-controlled, well-characterized, large-scale experiments simulating realistic thermal/mechanical loads to the reactor pressure vessel. The purpose of the OLHF project is to investigate lower head failure for conditions of low reactor coolant system (RCS) pressure (2-5 MPa) and prototypic temperature differences across the vessel wall ($\Delta T_W$) of 200°K to 400°K.

## Research Progress

The previous NRC-sponsored research investigated the condition of high RCS pressures and small $\Delta T_W$. Low RCS pressure is chosen because of the desire to use the data to develop models for assessing accident management strategies involving RPV depressurization. Pressure transient data is useful in assessing the effect of water injection as part of an accident management strategy. Prototypic $\Delta T_W$ is important because of the important need to provide data where stress redistribution occurs in the vessel wall (as a result of decreasing material strength with temperature). The OLHF experiments have been performed using 1 to 4.85 linear scale models of a typical pressurized water reactor (PWR) lower head. Figure 1 is a picture of the OLHF test assembly. The test vessel is geometrically scaled to a typical PWR reactor to preserve attributes of the membrane stress.

The test vessel consists of a 91.4 cm inner diameter, nominally 70 mm thick SA533B1 steel hemisphere welded to a 45 cm upright cylinder assembly closed off on top by a blank flange. The vessel is heated from within with a unique induction-heated graphite-radiating cavity. Large $\Delta T_W$ is achieved by increasing (with respect to geometrical scaling) the wall thickness and leaving the wall non-insulated. The membrane stress is preserved by increasing the test pressure by a factor corresponding to the wall thickness distortion (RW), i.e., $P_{test}=RW \bullet P_{RCS}$. The prototypic material for U.S. PWRs, SA533B1, was used to preserve material behavior.

Four integral tests were performed as part of the OLHF project. OLHF-1 and OLHF-2 are performed at 2 MPa and 5 MPa RCS pressure respectively. OLHF-3 examined the effect of pressure transients as the vessel wall passed through the ferrite-austenite phase transition region while the RCS pressure increased from an initial pressure of 2 MPa to a transient upper plateau of 5 MPa. The final test, OLHF-4, examined the effect of penetrations on vessel failure and was also tested at 2 MPa RCS (similar to OLHF-2). The bottom 120° of the test vessel was uniformly heated with a heat-up rate of 12°K/min for all tests.

Figure 1. The photograph is a post-test view of the OLHF-2 vessel on the test pad.

Two extra vessels were fabricated from the SA533B1 steel from which samples were prepared for material property testing. This material underwent essentially the same heat treatment and work history as actual test vessels to minimize variability in material properties. Five tensile tests were performed at high temperatures (925°K to 1,275°K) and 21 creep tests were performed at about 75 percent and 95 percent of yield stress at high temperatures (925°K to 1,275°K). Two replicate creep tests were performed to provide a means of assessing the reproducibility of test results. Supplemental tests were also performed by the French Atomic Energy Commission (CEA[1]) of France to extend and/or verify the database. The results of these tests are used to construct a constitutive model for implementation into structural analysis models.

[1] Commissariat à L'Energie Atomique

Independent numerical simulations were performed for the OLHF-1 test by all OECD partners in an international benchmark activity. Using finite element methods or analytical calculations together with material property data and test data for geometry description and boundary conditions, participants evaluated the time to failure or the value of damage parameters at the experimental failure time as well as variations with time of several mechanical variables. Overall, numerical models correctly estimate the timing, mode, and location of vessel failure. However, more work is needed in developing models of the crack opening and crack propagation to assess the size of the breach.

Simple "engineering" methodologies are required in severe accident codes (e.g., MELCOR, SCDAP/RELAP5, and MAAP4) that model the full sequence of events that occur in a core melt accident. It has been demonstrated (NUREG/CR-5582) that the creep-based methods utilizing the semi-empirical lifetime rule nominally correlate both the time for onset of creep and failure times observed in the NRC-sponsored LHF tests. These models have been assessed against the experimental data where large through-wall temperature gradients are important, resulting in significant redistribution of internal stress from the hot inside surface to the cooler outside surface of the lower head.

## Summary of Project Accomplishments

A summary is provided for the entire project. The accomplishments will be presented in terms of the three key program elements: integral experiments, material characterization, and model development and validation. Material characterization is the link between integral experiments and modeling.

**Key observations from integral experiments**

(1.) Large temperature differential leads to failure at higher inside wall temperature.

Comparison of the results of OLHF-1 and earlier tests (i.e., LHF-7) demonstrates the importance of stress redistribution on vessel deformation. Both tests performed with large through-wall temperature differences (OLHF-1 and OLHF-2) showed signs of non-linear deformation at higher temperatures than similar tests with small through-wall temperature differentials (LHF). The LHF tests showed signs of non-linear deformation when the inside surface was well below the yield stress. For the conditions of the OLHF tests, the onset of nonlinear deformation occurs as more than 10 percent of the vessel wall exceeds the yield stress. Failure in the OLHF tests occurred at higher temperatures than in corresponding LHF tests (small temperature differential). Since penetration failure was governed by global vessel deformation, this was also true of the penetration test.

(2.) Failures are typically localized.

Both the LHF and OLHF experiments as well as failure of reactor vessel retention (FOREVER) experiments revealed that the initiation of the failures is typically local. Failure was found to initiate at the location of maximum membrane to yield stress ratio. For OLHF experiments, since the load is carried by the cooler outer region of the wall, the stress ratio is evaluated at the external wall temperature. For the case of uniform temperature distribution, the crack initiates in the thinnest region because the location corresponds to maximum membrane stress. The crack initiates at the highest temperature regions for the case of non-uniform temperature distribution because yield stress is minimum at the highest temperature region.

Following this, most of the tests exhibited a localized propagation. Nevertheless, re-pressurization at elevated temperature can lead to rapid onset of non-linear deformation and failure and result in larger failure sites. This was observed in both OLHF-3 and LHF-5 tests.

It is important to note that the test results cannot be directly used to assess failure size for reactor cases because the rate of depressurization depends on the gas volume in the system, which is not scaled in the OLHF or the LHF tests. Furthermore, in the reactor case, molten corium ejection could enlarge the failure site through ablation.

(3.) Consistent global failure strain was observed.

The critical effective strain is often used as a failure criterion in modeling the vessel deformation in Severe Accident Codes. The critical effective failure strain for uniformly heated vessel without penetration was found to be ~30 percent. This value is consistent among the first three OLHF tests, and uniformly heated LHF tests.

Penetration failure has been found to occur as a result of global deformation of the lower head leading to failure at the weld vessel interface. Penetration failure occurs at much lower effective strain, i.e., 10 percent for OLHF-4 and 7 percent for LHF-4. In both experiments, failure initiated at the weld-vessel interface, which resulted in depressurization and termination of experiments. Direct application of the results to the reactor cases is questionable because understanding of the scaling effect of weld failure is currently lacking; furthermore, the effects of molten corium is not simulated in the OLHF experiments. Provisionally, one can assume that initiation of penetration failure due to global deformation occurs at a critical effective strain of ~10 percent. Metallurgical and material properties of weld are important topics that should be addressed for developing predictive model of penetration failure.

**Key observations from material characterization experiments**

The tensile and creep properties were measured for temperature up to approximately 1,300°K for LHF and OLHF steel. There is general consistency between the data from SNL and CEA and the external database established in the LHF program. The data fits (except elastic modulus) developed in the LHF program were used in OLHF analyses and numerical simulations.

However, a close examination of the data indicates that for future analysis of OLHF integral experiments it might be advisable to refit the data with more weight given to measured OLHF material properties. Fits giving equal weights to all data can be used for general assessment purposes. It is also important to note that comparisons between the SNL and CEA measurements indicate that there are laboratory-to-laboratory differences in the OLHF material data set. The effort and resources required to resolve such difference are likely to be quite high. At the moment it is perhaps best to acknowledge the difference as an irreducible uncertainty.

CEA has performed metallurgical characterization of failure sites of LHF and OLHF material. For high temperatures (> 1000°K), LHF material exhibits brittle failure whereas OLHF material exhibits ductile behavior.

CEA concluded that the difference could be attributed to the nearly 10 fold higher sulfur content in the LHF material as compared to the OLHF material although both materials conform to the specifications of SA533B1 steel. The effect of the ductile versus brittle behavior on critical strain, failure propagation, and final failure size should be assessed.

The ferrite to austenite phase transition region for the OLHF material has been determined to be between 1005°K and 1133°K using dilatometry. The tests also showed that there are no orientation effects although the grain structure does indicate the effect of cold work, i.e., finer grain near the inner and outer surfaces. The rate of vessel deformation diminished or reversed as the vessel wall passed through the phase transition from ferritic to austenitic steel. However, the effect of phase transition appears not to be as significant as once considered probably because only a portion of the wall is in the transition region at any instance of time and the induced stresses due to phase transition are of second order.

**Key Observations on Severe Accident Codes and Numerical Simulations in Model Development and Validation**

The OLHF project has produced a unique and well-qualified data set for code validation.

A benchmark exercise based on the OLHF-1 test data has been conducted by the OLHF participants. Different failure criteria have been used (damage or strain) by the participants with no consensus or convincing argument for any preference. Generally, the predicted failure times calculated by participants agree reasonably well with the test data. All the models, irrespective of their complexity, gave reasonable prediction of the failure time. It should be noted that failure time is not a rigorous measure of model predictability because the time to non-linear deformation is long compared to the time interval between the initiation of non-linear deformation and vessel failure. When referenced to the onset of non-linear deformation, there is still considerable spread in the predicted time to failure.

The OLHF-4 (penetration) vessel provides a snapshot of the deformed vessel head prior to failure. Since the OLHF-4 vessel appears nearly symmetric, this provides some evidence that early deformation in the vessel head is axi-symmetric. Calculation performed by Finnish participants showed that penetration behavior in OLHF-4 could be adequately represented by a 2-D simulation.

On the other hand, location and extent of failure site must be determined by 3-D modeling. The OLHF tests as well as LHF tests demonstrate that rather small asymmetries in temperature or wall thickness could lead to large asymmetries in the overall vessel deformation. Therefore, a 3-D model would be needed to completely characterize the vessel deformation and to predict the location and extent of the failure site. The propagation of the failure and final size of the failure is still an issue; 3-D calculations by CEA, presented in the OLHF 2002 Seminar, show promising results.

The simple SAC models that were evaluated in this program proved quite adequate in predicting vessel failure in the OLHF tests. Various failure criteria, damage functions, and strain rate equations were assessed. For the conditions of these tests, the accuracy of the various models in predicting the test failure times appears to lie within the spread of results obtained by the Finate Element Analysis (FEA) benchmark analysis. This would support the use of such simple models for the purpose of severe accident analysis. However, the correlation for LMP parameters should be based on OLHF property data, as those based on LHF data predict earlier failure.

## OLHF Seminar

An OLHF seminar, sponsored by OECD, was held in June 2002 to provide an in-depth review of the project technical capabilities, results, and analyses. Progress in the areas of material characterization, finite element model improvements, modeling vessel penetrations, crack propagation, and simplified models for severe accident analysis, is found in the Proceedings.

## Planned Activities

This completes the OLHF experimental activities; the final report will be published as an OECD report.

The data from these tests has been well-qualified and well-archived for future reference. The data is organized into Microsoft Excel spreadsheets with numerous macros for visualization and simple analysis of the data. This project has produced a database that has become an international standard for assessing vessel deformation and failure models.

NUCLEAR ENERGY RESEARCH INITIATIVE

## The Secure Transportable Autonomous Light Water Reactor—STAR-LW (IRIS Project)

Primary Investigator: Mario D. Carelli, Westinghouse Electric Company LLC

Collaborators: University of California, Berkeley, USA; Massachusetts Institute of Technology (MIT); Polytechnic Institute of Milan, Italy

Project Number: 99-027

Project Start Date: August 1999

Project End Date: January 2003

### Research Objective

This program, currently known as the International Reactor Innovative and Secure (IRIS) project, has the objective of investigating a novel type of water-cooled reactor which can satisfy the Generation IV goals: fuel cycle sustainability, enhanced reliability and safety, and improved economics. The research objectives over the three-year program are as follows:

- First year: Assess various design alternatives and establish main characteristics of a point design.
- Second year: Perform feasibility and engineering assessment of the selected design solutions.
- Third year: Complete reactor design and performance evaluation, including cost assessment.

### Research Progress

The original four member, two-country team grew to 11 members in the first year of activity, and further expanded to 18 members from nine countries at the end of the third year, although the French Atomic Energy Commission (CEA [1]) withdrew at the end of the first year and the Japanese JAPC and MHI had withdrawn by the end of the second year. Two more organizations from Brazil, Eletronuclear, and Industrias Nucleares do Brazil, are considering joining the IRIS team. All the added team members work under their own funding and it is estimated that the value of their in-kind contributions in the second year was about $8M, which grew to approximately $12M in the third year. Four universities (University of Tennessee, Ohio State University, University of Michigan and Iowa State University) and two laboratories (Ames and Sandia) also became associated with the program through additional NERI programs and students projects. To date, 72 students have worked or are working on IRIS. By December 2002, 51 IRIS-related graduate theses will have been prepared or are in preparation, and 28 students will have graduated with M.S. or Ph.D. degrees.

The large increase in additional effort, not envisioned in the proposal submitted, has allowed the researchers to significantly exceed their original objectives and to change the outlook for IRIS from a long-term R&D project to a commercially viable design with a deployment target date in the next decade. Several interactions have taken place with NRC at various levels (commissioners, staff, and ACRS) and IRIS pre-application licensing was formally initiated in October 2002.

In response to requests from utilities, IRIS site layout and site bounding information were provided to the ESP (early site permit) program.

The IRIS conceptual design was completed. A summary of program highlights follows.

- The changed emphasis towards a competitive, early deployable reactor has led to two major changes in the IRIS design. First, the reference IRIS size was set at 1,000 MWt (~ 335 MWe), although the same design configuration covers the 100 to 335 MWe range with only modest changes in dimensions. Second, the core design now features a 4.95 percent enriched $UO_2$ fuel in a 17x17 square array assembly (a 15 x 15 assembly is also possible), very similar to standard Westinghouse pressurized water reactor (PWR) assemblies. Since this fuel has an expected straight burn lifetime of slightly more than four years, with a burn-up of approximately 40,000 MWd/t, it presents no licensing issues. The IRIS core is designed to accept various configurations (8-year straight burn, 8-10 percent fissile $UO_2$ and MOX fuel;

[1] Commissariat à L'Energie Atomique

4-year straight burn 4.95 percent enriched $UO_2$; two and three batch 4.95 percent enriched $UO_2$). The latter are for initial deployment, while the former can be considered for future reloads.

- The IRIS vessel (see Figure 1) includes eight helical steam generators, eight "spool-type" pumps, and pressurizer and internal shields. Six different steam generator designs were evaluated and the Ansaldo helical design was chosen both for its performance and for the fact that it had already been extensively tested in a 20 MWt mockup. The fully internal pumps are based on a design developed for chemical applications. They can be operated in a high-temperature environment and can have large coastdown and run-out capabilities, but have to be qualified for nuclear applications. The pressurizer is of the steam type and the ratio of its volume to reactor power is favorable in being much larger than loop PWRs, thereby allowing very smooth pressure control. The internal shields fill very conveniently into the annular space between the core and the vessel and they reduce the radiation field at the vessel outer surface to the order of $10^{-4}$ Sv/hr. This has very positive implications for operational and maintenance doses, for long vessel life, as well as for decommissioning and disposal (the "cold" vessel can act as a sarcophagus for the whole reactor internals minus the fuel).

- The concept of "safety by design" (to physically prevent accidents from occurring rather than coping, by active or passive means, with their consequences) has been developed and articulated in detail and its implementation has widely exceeded expectations. Not only are large loss of coolant accidents (LOCAs) eliminated from occurring, as can be expected with all integral designs, but a patented containment design has also practically eliminated small and medium LOCAs as a safety concern. In fact, the core remains fully covered for an extended period of time, several days and possibly weeks, without any safety injection or water make-up. This is made possible by a design that thermo-hydraulically couples the vessel and the containment so that within the first hour after the pipe breaks, the pressures inside the vessel and the outside pressure in the containment equalize, thereby canceling the differential pressure that drives the coolant across the break. Also, the IRIS vessel has no penetrations at and below the core region, and it sits in an open cavity that extends above the core level. Suppression pools are located in the containment and they can double as gravity makeup, even though they are not needed. Decay heat is removed by four diverse (three independent) systems: eight steam generators, four natural circulation heat exchangers located outside the containment, and surface (air and water) containment-cooling.

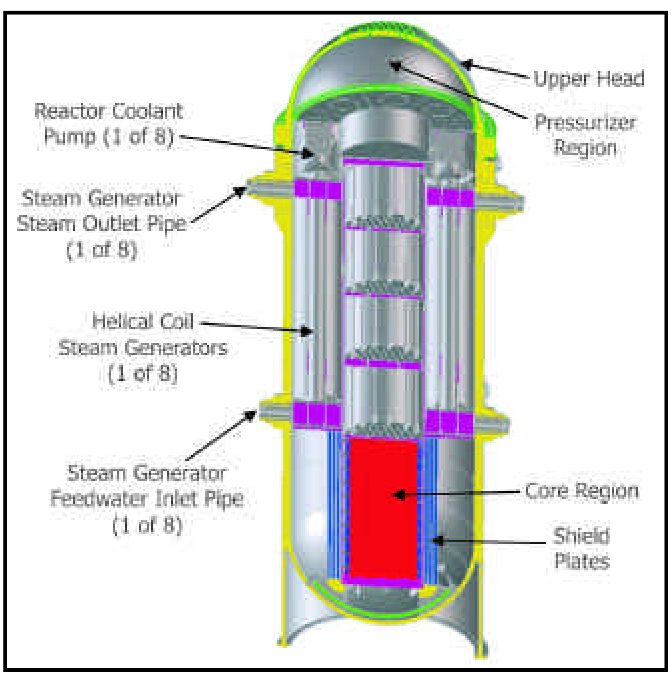



Figure 1. The diagram shows the assembly of the IRIS NSSS.

Loss of flow accidents (LOFAs) have no significant consequences because of the pumps' characteristics and redundancy, as well as the substantial degree of natural circulation. Steam generator tube rupture accidents have lower probability and more benign consequences since the tubes are in compression (with primary coolant outside) and are designed for zero internal pressure.

The conclusion is that IRIS is a water reactor design where primary coolant-related accidents are of no major concern, and of the eight Class IV accidents typically considered in the advanced passive designs safety analysis reports (SARs), only one (refueling accident) remains as a Class IV accident and even that, with a much lower probability. All the others either no longer apply, or can be reclassified at Class III or lower.

- An IRIS model using the RELAP5 code was completed to perform initial plant safety assessment and was verified in a preliminary steady state and transient qualification. All the relevant transient events and accidents typically reported in a Safety

Analysis Report were assessed, including (but not limited to) steam system piping failure, feed system piping failure, loss of offsite power, turbine trip, loss of flow, locked rotor, reactivity anomalies, steam generator tube failure, small break LOCA, and anticipated transients without scram.

In accordance with standard procedures, the system code (RELAP) is coupled with subchannel and neutronic analysis codes when required by the specific event considered. CFD analyses of selected portions of the pressure vessel have been performed to verify mixing phenomena in the IRIS system. For the analyses of small break LOCA, the strong coupling between vessel and containment during most of the event duration has required development of new approaches for the system analyses. While different solutions have been explored, a thermal-hydraulic coupling of RELAP (for reactor coolant system analysis) and GOTHIC (for containment analyses) was identified as the most promising approach and used in the analyses.

Models and results of the analyses have been collected in a preliminary plant safety assessment document to be submitted to the NRC as part of the IRIS pre-application review. Probabilistic Safety Assessment (PSA) analyses have been initiated, with a preliminary assessment of events and fault trees.

- Substantial work has been completed to support the IRIS goal of a 48-month interval between maintenance shutdowns. This, coupled with the core lifetime of about four years without refueling, will yield very high capacity factors and significantly reduce the operating and maintenance (O&M) costs. A previous effort was performed by MIT to investigate the feasibility of extending the maintenance interval in a commercial PWR from 18 to 48 months. A total of 3,743 maintenance items were identified for the 18-month cycle, 1,206 to be performed on-line and 2,537 off-line during the scheduled outage. The MIT study showed that most of the 2,537 off-line items could be deferred to 48 months or be performed on-line. Only 54 items in various categories (e.g., relief valves, motor operated valves, pumps) remained outstanding, as they still required an 18-month interval.

  Building on this study, the unresolved items were examined for their applicability to IRIS (e.g., pump oil lubrication obviously does not apply to the reactor coolant lubricated internal spool pumps). Only seven items in five categories were finally identified as still outstanding impediments to a 48-month maintenance interval in IRIS. None of these are considered showstoppers and work is in progress for their resolution. An additional category was identified for items that could be tested on-line, but would require a reduced power level for the test.

- As part of the ESP input, two potential arrangements of multiple IRIS modules were identified: one (1,000 MWe total) consisting of three modules "in a string" with staggered construction start and one (1,350 MWe total) consisting of two twin units where each unit has two modules sharing almost all auxiliary systems.

- A market analysis and preliminary top-down cost estimate was performed, confirming the competitive attractiveness of IRIS, both in developed and emerging countries. The total cost of electricity was on the order of $0.03/kWh, with a best estimate of $0.0285/kWh.

- The necessary testing program to confirm the operational and safety characteristics of IRIS is being outlined. A preliminary assessment is underway, which includes preparation of PIRTs (Phenomena Identification and Ranking Table), identification of required tests, specification of parameters, assessment of similitude analyses, preparation of test plans, and identification of test facilities.

- Risk informed regulation is being assessed as an option for the IRIS licensing, with the objective of demonstrating that IRIS can achieve the stated Generation IV goal of eliminating the need for offsite emergency response planning.

### Planned Activities

IRIS development does not end with the conclusion of the NERI three-year program. The IRIS consortium is proceeding with detailed design and analyses, the NRC licensing process has been initiated, and the ESP program will continue with IRIS participation. In reality, IRIS development will proceed on a higher scale to fulfill the objective of producing a successful commercial entry in the next decade.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Monitoring and Control Technologies for the Secure Transportable Autonomous Reactor (STAR)

Primary Investigator: Hussein S. Khalil, Argonne National Laboratory

Collaborators: Lawrence Livermore National Laboratory (LLNL); Texas A&M University

Project Number: 99-043

Project Start Date: August 1999

End Date: September 2002

## Research Objectives

A new reactor and fuel system concept designated as the Secure Transportable Autonomous Reactor (STAR) has been proposed for meeting the needs of developing countries for small, economical nuclear power stations while at the same time addressing proliferation concerns. This NERI project supports this goal through development of operations-monitoring, and control and remote surveillance strategies that exploit the passive safety and autonomous operation attributes of the STAR plant. It also entails development and demonstration of advanced technologies for implementing these strategies to assure operational reliability and security of nuclear materials.

Specific objectives of the research are to simplify active control and safety protection systems; minimize reliance on on-site operating staff; and assure high levels of operational safety, reliability, and facility security. Research tasks include evaluating the ability of candidate STAR plants to operate autonomously with minimal reliance on active control for load adjustment and burnup reactivity compensation, identifying design and operating features that enhance operational autonomy and passive safety, developing simplified control strategies on the basis of the passive plant response, and developing and demonstrating computer-based technologies for remote monitoring of operational and safeguards information at centralized surveillance facilities.

Although proposed in the framework of the development effort for the STAR system, the research addresses issues of fundamental importance to the operation of passively safe and autonomous plants. Resolution of these issues will increase the immunity of passively safe plants to operator and control system errors and provide a technical basis for reducing the cost of plant control and safety protection systems.

## Research Progress

Accomplishments in the first two years of the project include the following:

<u>Autonomous Operability of STAR Designs:</u> Design options were identified for achieving autonomy of operation in the lead-bismuth eutectic (LBE) cooled STAR-LBE concept (see Figure 1). The approach for increasing autonomy focuses on the use of inherent properties of mechanical, hydraulic, thermal, and neutronics reactor systems, which are determined by the choice and arrangement of reactor materials. One consequence of the enhanced use of intrinsic feedbacks is reduced need for plant actuators.

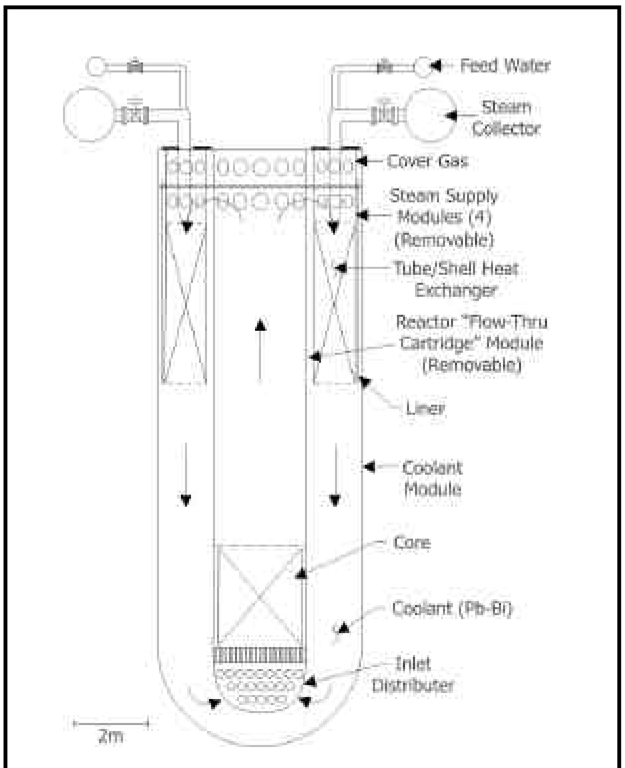



Figure 1. The diagram shows a STAR-LM simplified, modular, small reactor featuring a flow-through fuel cartridge.

Buoyancy-driven natural circulation eliminates primary pumps, temperature reactivity feedbacks replace rods for changing power, and internal conversion eliminates rods otherwise needed for compensation of reactivity excess at beginning of cycle. Substantial flexibility exists in the form of fuel type, core layout, reactor steel selection, primary loop layout, steam generator design, and selection of water-side equipment. It was found that inherent feedbacks can be used to make the STAR-LBE plant virtually self-regulating with respect to load change, reactivity control with burnup, and unprotected upsets. Startup, however, was found to require the insertion of rod worth to overcome reactivity associated with a negative power coefficient. The negative coefficient is dictated by stability requirements and is a technical specification. Other than needing a control system to deliver this worth, inherent feedbacks can provide for operation that is virtually free of the need for active control elements. Moreover, the analsis of reactivity effects for STAR-LBE with metallic fuel showed that these are no combinations of operator or control system errors that could cause reactor temperatures to exceed safe limits.

The possibility of using reactivity feedbacks to maneuver power and avoid the use of control rods was also explored for a natural-circulation boiling water version of the STAR concept (STAR-BWR). The first step in designing a STAR-BWR that does not use control rods to accomplish load following is to demonstrate that the desired range of steady-state powers is achievable. This has been the focus of the current effort. The base case 100-MWe plant used conventional uranium oxide fuel and was found to be capable of reaching a minimum power of only 82 MWe during load following. The alternative 100-MWe plant was capable of reaching 37.5 MWe. The alternative, however, assumes a very high fuel conductivity, indicative of metallic fuel, and a doubling of the ratio of the void coefficient to the temperature coefficient. Additional study of the thermal and neutronics aspects of a BWR and its fuels would thus be necessary to determine what is achievable in practice. Moveover, dynamic behavior and stability implications of reliance on passive control and natural circulation cooling should be examined in future work.

<u>Remote Monitoring System Design:</u> A design for a remote monitoring system was developed to meet anticipated security and operational monitoring requirements for a STAR plant. In this system, key plant security signals are acquired, digitized, encrypted, and sent via the communication system to the remote monitoring site. Security requirements were defined assuming all persons with normal access to the plant may be a threat. Accordingly, the security requirements start at the sensors and extend through the communications system. Many of the sensors have dual use for security and operation (e.g., reactor temperatures and flux levels), while others are dedicated to security (e.g., motion or volume sensors, video cameras, and so forth). This data is transmitted to the remote site with a sampling rate appropriate to the data, but no more frequently than once each second. This data would be monitored at the remote site essentially in real time and would be stored for trending displays and additional analyses. Features to thwart potential attempts to subvert the monitoring system include limiting access to system components and use of a robust combination of tamper detecting devices and information analysis capabilities based on "machine intelligence" techniques.

Key features of the monitoring system design have been implemented in cooperation with Texas A&M University. A satellite-based network was established to allow remote observation of the Texas A&M Nuclear Science Center Reactor, and to provide integrated voice video and data transmission between LLNL and the reactor.

<u>Demonstration of Remote Reactor Monitoring:</u> A capability was developed and implemented for remote monitoring of the Texas A&M University Nuclear Science Center Reactor (NSCR). This capability uses standard hardware components and a software package (LabVIEW) available from National Instruments Co. for data and image acquisition, information analysis and storage, and remote monitoring of this information on the Internet using either browser or LabVIEW software. The new hardware and software were installed to provide a capability for Internet- or satellite-based viewing of the NSCR main control room console, the fuel and water system temperatures, the reactor log and linear power readings, control rod positions, and Facility Air Monitoring System data. In addition, a video-based surveillance system has been designed and implemented for remote viewing of the NSCR core. This surveillance system is illustrated in Figure 2. The required video equipment was procured and interfaced with the LabVIEW software for display at the monitoring site.

In addition to simplifying the control and safety protection systems, the STAR plant design and the proposed approach to plant control promise to allow uninhibited deployment of computer-based operations

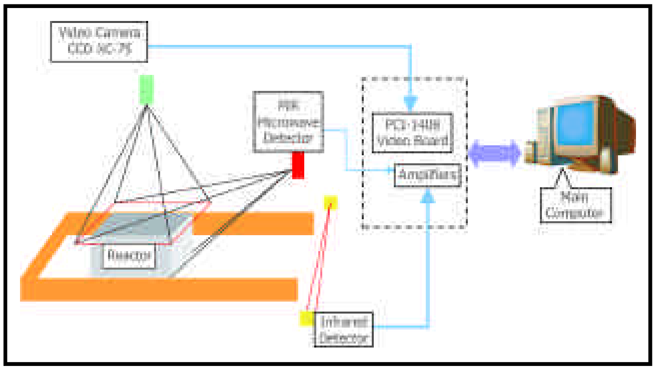



Figure 2. The diagram shows the video monitoring system implemented for the NSCR at Texas A&M University.

support systems that exploit modern digital-based computing and information management technologies. Key roles (functionality) identified for these technologies are to (a) generate, substantiate, and graphically display sensor information required to describe the state of plant systems, and (b) exploit this information to maximize plant availability, optimize maintenance activities including those requiring the dispatch of specialists to the plant site, and assist on-site operators to cope safely and effectively with potential upsets. Software reliability is the key challenge identified until now for the deployment of these digital technologies.

To address this challenge and provide the needed functionality, operator support systems should satisfy key requirements including inherent portability, ability to accomodate unforeseen events and amenability to generic verification and validation. These requirements could be satisfied by employing a first-principles modeling approach augmented by the use of measured data. This approach would overcome the limitations of hard-wired course/symptom methods that are inherently plant- and process-specific.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Risk Informed Assessment of Regulatory and Design Requirements for Future Nuclear Power Plants

Primary Investigator: Stanley E. Ritterbusch, ABB-Combustion Engineering

Collaborators: Sandia National Laboratories; Idaho National Engineering & Environmental Laboratory; Massachusetts Institute of Technology; North Carolina State University; Duke Engineering Services; Eagan & Associates

Project Number: 99-058

Project Start Date: August 1999

Project End Date: December 2002

## Research Objective

Research under this project addresses the barriers to long-term use of nuclear-generated electricity in the United States. Studies being performed by the Electric Power Research Institute on the cost of coal, gas, and nuclear-generated electricity have identified that to be competitive, the cost for the nuclear option would have to decrease to the range of $0.03/kilowatt-hour over the next two decades. Correspondingly, the total plant capital cost of a typical Advanced Light Water Reactor (ALWR) would have to decrease by about 35 percent to 40 percent and the construction schedule would have to be shortened to about three years.

## Research Progress

Shortly after initiating this project, team members established the principal strategies required to achieve the project's cost-reduction goals. It was agreed that a very basic and significant change to the current method of design and regulation was needed. That is, it was believed that the cost-reduction goal could not be met by fixing the current system (i.e., an evolutionary approach) and a new, more advanced approach for this project would be needed. It is believed that a completely new design and regulatory process would have to be developed-a "clean sheet of paper" approach. This new approach would start with risk-based methods, would establish probabilistic design criteria, and would implement defense-in-depth only when necessary to (1) meet public policy issues (e.g., use of a containment building no matter how low the probability of a large release) and (2) address uncertainties in probabilistic methods and equipment performance. This new approach is significantly different from the Nuclear Regulatory Commission's (NRC's) current risk-informed program for operating plants. For the new approach, risk-based methods are the primary means for assuring plant safety, whereas in the NRC's current approach, defense-in-depth remains the primary means of assuring safety.

The primary accomplishments in the first year (Phase 1) of this project include

(1) The establishment of a new, highly risk-informed design and regulatory framework;

(2) The establishment of the preliminary version of the new, highly risk-informed design process;

(3) Core damage frequency predictions showing that, based on new, lower pipe-rupture probabilities, the design of the emergency core cooling system equipment can be simplified without reducing plant safety; and

(4) The initial development of methods for including uncertainties in a new integrated structures-systems design model.

Under the new regulatory framework, options for the use of "design basis accidents" were evaluated. Whereas, in the current regulatory process, the design basis accidents and their evaluation are primarily deterministic, it is expected that design basis accidents would be an inherent part of the Probabilistic Safety Assessment for the plant and their evaluation would be probabilistic.

Other first year accomplishments include

(1) The conversion of an NRC database for cross-referencing NRC criteria and industry codes and standards to Microsoft 2000 software;

(2) An assessment of the NRC's hearing process,

which concluded that the normal cross-examination during public hearings is not actually required by the U.S. Administrative Procedures Act;

(3) The identification and listing of reliability data sources; and

(4) Time spent interfacing with other industry groups [e.g., Nuclear Energy Institute (NEI) and International Atomic Energy Agency (IAEA)] and NRC at workshops for risk-informing regulations.

The foregoing tasks were continued during the second year (Phase 2) of this project. In addition, Westinghouse signed an agreement with the Korea Power Engineering Company (KOPEC) for their participation in this project. Major benefits of this cooperation include the Korean design and operating experience that is brought to the project, and the completion of a relevant work scope by KOPEC personnel. This agreement was reviewed and approved by other project participants as well as the Department of Energy.

The major accomplishments during the second year consisted of

(1) Issuance of the final report for Subtask 1.1, "Identify Current Applicable Regulatory Requirements [and Industry Standards]";

(2) Issuance of the final report for Subtask 1.2, "Identify Structures, Systems, and Components and Their Associated Costs for a Typical Plant";

(3) Extension of the new, highly risk-informed design and regulatory framework to non-light-water-reactor technology;

(4) Completion of more detailed thermal-hydraulic and probabilistic analyses of advanced conceptual reactor system/component designs;

(5) Initial evaluation and recommendations for improvement of the NRC design review process; and

(6) Initial development of the software format, procedures, and statistical routines needed to store, analyze, and retrieve the available reliability data.

In Phase 2 of the project, several reports and documents were produced as planned. Final reports for Subtasks 1.1 (regulatory and design criteria) and 1.2 (costs for structures, systems, and components) were prepared and issued. A final report for Subtask 1.3 (Regulatory Framework) was drafted with the aim of issuing it in Phase 3 (Year 3). One technical report was produced for Subtask 1.4 (methods development) and two technical reports were produced for Subtask 1.6 (sample problem analysis). An interim report on the NRC design review process (Subtask 1.7) was prepared and issued. Finally, a report on Subtask 2.2 (database weaknesses) addressed the initial development of a new database to track reliability data.

During the third and final year (Phase 3) of this project, work was completed on Subtasks 1.3 (regulatory framework), 1.6 (sample problem analysis), Subtask 1.7 (regulatory analysis), Subtask 1.8 (industry and NRC coordination), and Subtask 2.3 (reliability data improvements). Also during the third year, more detailed thermal-hydraulic and probabilistic analyses of the advanced conceptual designs were completed. The results of these efforts will be documented in subtask reports and in the final report for this project. The regulatory framework was updated to reflect the extension of new risk-informed methods to non-LWRs (Subtask 1.3), and supporting analyses for a pebble-bed reactor design were completed (Subtask 1.6). Subtask 1.6 also covered additional thermal-hydraulic analysis for the current generation LWR loss of coolant accident. Work on software to facilitate access to reliability data was completed in Subtask 2.3.

## Planned Activities

Final subtask reports for the subtasks completed in Phase 3 (indicated above) will be issued. Also, the Phase 3 Annual Report and the project's Final Report covering all three years will be issued.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Demand-Driven Nuclear Energized Module

Primary Investigator: Gary T. Mays, Oak Ridge National Laboratory (ORNL)

Project Number: 99-064

Project Start Date: August 1999

Project End Date: September 2002

## Research Objectives

The Nuclear Energizer Module (NEM) is a reactor concept designed to take advantage of newly developed graphite foam, which has enhanced heat transfer characteristics and excellent high-temperature mechanical properties and will provide an inherently safe, self-regulated source of heat for power and other potential applications.

The principal objectives of the project are to (1) develop a reactor concept with a target power level of 500 kW(th) that is naturally load following, inherently safe, optimized via neutronic studies to achieve near-zero reactivity change with burn-up, and proliferation-resistant; (2) prepare and appropriately characterize the physical properties of the graphite foam; (3) conduct irradiation studies of the graphite foam to determine any effects on structure, dimensional stability, thermal conductivity, and thermal expansion; (4) simulate the overall performance of the reactor concept in terms of operations, safety, stability, and thermal characteristics; and (5) develop a physical model using the graphite foam and electric heaters to benchmark computer models. The general application targeted for this concept is a design that is easily deployable to supply power to remote and/or harsh environments.

## Research Progress

This project has four major tasks, in the following areas:

- Neutronics
- Materials testing and evaluation
- Simulation and thermal analysis
- Power conversion

A brief summary of progress in each of these areas follows.

Neutronics Analyses: The reactor core consists of a right circular cylinder of graphite foam impregnated with uranium carbide. The radius of the cylinder is 75 cm. The cylinder is divided into two zones: a central unpoisoned region having a radius of approximately 13 cm and an outer annular region in which the foam is impregnated with cadmium at a Cd density of 86 milligrams per $cm^3$. The $C/^{235}U$ ratio is 39. The uranium [20 percent enriched U; critical mass of about 4 Metric Tons of Uranium (MTU)] exists as $UC_2$. During normal operation, the cadmium is relatively innocuous (the reactor has a fast spectrum). However, should there be ingress of water to the core, the cadmium acts as an effective neutron poison causing the reactor to be subcritical. The temperature coefficient of the fueled-foam is expected to be between -0.2 and -0.3 pcm/°K.

The core is surrounded by a graphite reflector (i.e., normal graphite, not graphite foam). Uranium-impregnated foam would be encased in a "super-alloy" steel. The reflector/clad interface temperature is expected to be limited to 900°K. The reflector thickness (considering reactor weight, size, and cost of fuel) is approximately 30 cm. Shutdown/startup of the reactor would be achieved by control elements in the radial reflector, outside the core clad.

Materials Testing: Four capsules, each with three foam samples plus a SiC temperature monitor, were irradiated in the High Flux Isotope Reactor at ORNL. Two capsules were used for the in-plane samples, and the other two were used for the out-of-plane samples. One in-plane and one out-of-plane capsule received an irradiation dose of 2.6 displacements per atom (dpa); while the other in-plane and out-of-plane capsule received an irradiation dose of 0.3 dpa. The location of the capsules within the hydraulic tube was such that the variation of the neutron flux was less than 15 percent from capsule to capsule.

Evaluation of the SiC temperature monitors allows the determination that the actual irradiation temperature of the capsules was approximately 820°C; this temperature is considerably higher than the planned irradiation temperature of 600°C. Observation of irradiated samples under a Scanning Electron Microscope (SEM) showed no apparent changes or damage, i.e., samples conserved their structural integrity during irradiation.

Thermal conductivity measurements and annealing studies of irradiated samples showed that thermal conductivity decreased as the irradiation dose increased. This effect is consistent with typical results for graphite samples. Annealing of irradiated foam samples to 1,000°C and 1,200°C lead to recovery of their thermal conductivity. Figure 1 shows the results of the thermal conductivity measurements as a function of the measurement temperature, for sample OP-9, irradiated at 2.6 dpa.

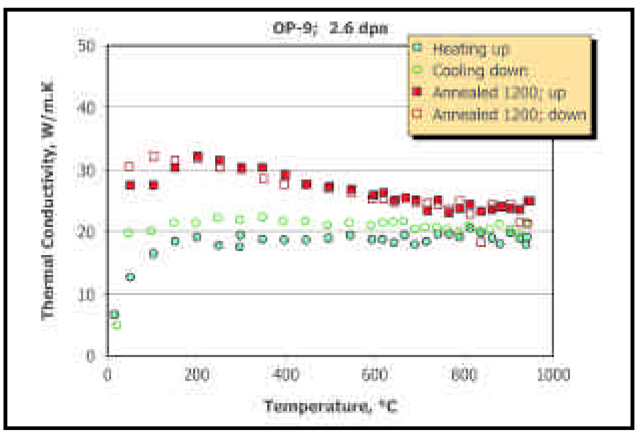



Figure 1. The graph represents thermal conductivity measurements of sample OP-9 after irradiation and after annealing to 1,200°C.

<u>Simulation and Thermal Analysis:</u> A variety of computations have been performed to assess the temperature distributions for candidate reactor configurations. Computations indicate that for the target 500 kW(th) fission heat generation, cooling on the top and bottom circular boundaries should be considered in addition to the cylindrical surfaces presently chosen. Heat removal at the core/reflector boundary instead of at the reflector's external boundary may also be required.

Two-dimensional computations of temperature distributions inside the core and reflector for gas cooling are being performed for the present configuration of heat removal only at the reflector's external cylindrical surface, heat removal only at the core/reflector's internal cylindrical surface, and heat removal through the bottom and top circular surfaces in addition to the cylindrical surfaces.

<u>Power Conversion:</u> The reference thermal power conversion system consists of the heat transfer geometry surrounding the nuclear core cladding and the closed Brayton cycle components that convert the thermal power generated by the reactor into electrical power. Heat transfer fins are necessary to increase the effective heat transfer area of the core clad surrounding the nuclear-fueled region. The fins will be made of the same material, a super-alloy steel as the core clad, and the clad surrounding the reflector will serve as an outer shroud to contain the cooling flow. Based on optimization studies, the fin geometry is as follows: fin height of 10 mm, fin width of 5 mm, and fin spacing of 10 mm. Using this cooling geometry, the working fluid pressure drop in the finned area is less than 1 percent.

The working fluid of the recuperated closed Brayton cycle system chosen is a mixture of helium and xenon, with a molecular weight of 40 (HeXe40). If pure helium were chosen as the working fluid, the relatively high thermal conductivity would result in the recuperator having a minimal necessary heat transfer area as compared to other gases, thereby minimizing the overall volume envelope of the system. However, pure helium also has a high specific heat that would limit the temperature change and therefore the pressure ratio across a stage. This stage pressure ratio limit would increase the number of turbine and compressor stages and result in complicated turbomachinery. Thus, a compromise is necessary between the small recuperator and complicated turbomachinery for pure helium and the larger recuperator with simple turbomachinery of HeXe40. Closed Brayton cycle turbomachines have been successfully designed and tested for space applications using HeXe40 in the electrical power range of interest (~100 kWe). Therefore, the HeXe40 turbomachinery design was chosen. Because the working fluid is clean and inert, relatively long full power operating intervals of a decade can be expected.

Because of core/clad interface temperature limitations, the turbine inlet temperature has been specified as 900°K. Since a terrestrial location is postulated for this reactor concept, a compressor inlet temperature of 310°K is specified. The ultimate heat sink of water, forced convection air, and natural convection air were analyzed. While water would result in the smallest size heat-rejection heat exchanger and perhaps a lower compressor inlet temperature, the presence of water vapor near the active core during an upset transient is a nuclear criticality consideration. A natural convection heat sink design would require an air chimney and have a large heat rejection heat exchanger. Thus, for simplicity, the forced convection air design is chosen as the ultimate heat sink.

Recuperator effectiveness is a measure of the actual heat transferred to the maximum possible from the hot side to the cold side of the recuperator. A reasonably achievable effectiveness is 95 percent which is within the existing industrial capabilities for manufacturing.

The system pressure drop is defined as the sum of the individual component fractional pressure drops. The component fractional pressure drop is defined as the actual pressure loss divided by the absolute working pressure of the component. This system fractional pressure drop determines how much of the turbine output is absorbed as parasitic pressure loss. A reasonable and realistic value of 5 percent is chosen, where 1 percent is budgeted for the core heat transfer fins and 4 percent for the remainder of the closed Brayton cycle. In order to keep the system as uncomplicated as possible, the compressor will not be intercooled.

Industrial firms under contract to NASA have designed HeXe40 turbomachinery appropriate for the electrical power range of interest here that use a radial flow design, which has been selected for this application. The compressor discharge pressure is 1.48 MPa (14.5 atm or 200 psig). This pressure, the highest in the system, is relatively low and results in a simple pressure containment boundary. A reasonable compressor polytropic efficiency of 85 percent and a turbine polytropic efficiency of 90 percent are achievable for this size turbomachinery.

Because the NASA turbomachinery was initially designed to use an 80 percent effective recuperator, the pressure ratio across the compressor is 2.2. Using this pressure ratio of 2.2 with the above cycle parameters results in a cycle thermal efficiency of 27 percent with a HeXe40 mass flow of 4.33 kg/s. Thus, for the proposed core thermal power of 500 kW, 135 kW of shaft power is available using the existing NASA turbomachine design. With a generator efficiency of 95 percent, the electrical power resulting from using the existing turbomachinery design is 128 kW.

However, since a 95 percent effective recuperator is sized here, the pressure ratio across the compressor that results in the optimum cycle efficiency is 1.65. Using this pressure ratio with the above cycle parameters results in a cycle thermal efficiency of 30 percent with a HeXe40 mass flow of 6.28 kg/s. Thus, for the proposed core thermal power of 500 kW, 150 kW of shaft power is available. With a generator efficiency of 95 percent, the electrical power resulting from this design is 143 kW. The existing NASA turbomachine design would have to be modified to the lower pressure ratio, which should be easily accommodated.

## Planned Activities

The remaining activity involves the preparation of a final report documenting the research and analysis performed and describing the preconceptual design for this reactor system.

# Nuclear Energy Research Initiative

## Development of Advanced Technologies to Reduce Design, Fabrication and Construction for Future Nuclear Power Plants

Primary Investigator: Camillo A. DiNunzio, Framatome ANP DE&S

Collaborators: North Carolina State University; Massachusetts Institute of Technology; Sandia National Laboratories; Westinghouse Electric Company Nuclear Systems; Korean Power Engineering Company, Inc. (KOPEC)

Project Number: 99-077

Project Start Date: August 1999

Project End Date: November 2002

### Research Objectives

The goal of the Design, Procurement, Construction, Installation, and Testing (DPCIT) project team is to identify methods that can deliver a 40 percent reduction in capital cost and scheduling time for a future nuclear power plant. Given the starting point of the Advanced Light Water Reactor (ALWR) program's benchmark of $1,500 per kW installed, and a 60-month construction schedule, the targeted reductions translate into a power plant built for $900 per kW and 36 months from first concrete to fuel load. Ideally, these techniques and innovations could be demonstrated in near-term reactor plants and then incorporated into the planned Generation IV reactor plants, which are under conceptual development. Most of the innovations planned in this project should be applicable to most reactor technologies since the concepts under consideration are not linked to a specific reactor design. However, the proposed reductions will be demonstrated using pressurized water reactor technology.

### Research Progress

The DPCIT project represents a merger of information technologies and supply chain management principles with design and construction improvements. The project focused on processes, and along the way adopted the appropriate tools to execute the process. Figure 1 provides a visual image of the intent of the DPCIT project.

The project consisted of tasks performed over a three year period, 1999-2002.

**Year 1 Summary**

During the first year of project activities (1999-2000), the project team identified initial strategies for reducing capital costs and scheduling. A key insight gained by the team is that back-fitting new technologies into pre-existing designs does not offer as significant a payoff as working with new designs as they are being created. This favors the Generation IV advanced nuclear plants since these plants are in the process of incorporating the kinds of improvements this project has examined, or there is the potential to incorporate these insights before their designs are finalized.

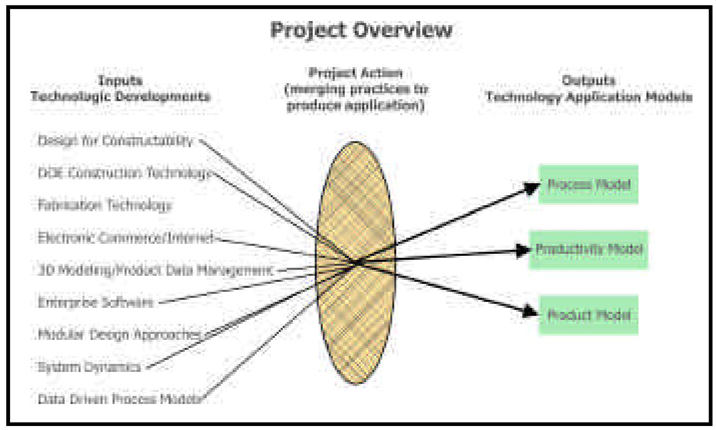



Figure 1. DPCIT project overview

**Year 2 Summary**

During the second year's activities, initiatives pursued included examining the potential impact of information technology on physical construction activities (the impacts of information technology on the design side of construction were examined in year 1); evaluating the potential for project management concepts to serve as advanced tools to help assure on-budget and on-schedule performance of a complex project such as a nuclear plant; and examining Cost Risk Modeling as a mechanism for assessing the viability of potential cost and schedule reduction techniques. Additionally, mechanisms were explored to remove excess conservatism and reduce error while shrinking the time between design and analysis activities, and methods were evaluated for removing excess seismic margins and simplifying containment construction.

**Year 3 Summary**

In year three, the final phase of this project, work focused on the following areas:

Containment and Structural Simplifications: Work continued on evaluating excess margins and simplifying structural systems. While reduced loads are likely to result by developing simpler alternative structural systems, a trade-off exists between the reduction of seismic margins and risk. Any decision regarding structural simplification should be focused on addressing this trade-off. During year 3 of this project, a decision support system (DSS) was developed for evaluating this trade-off. A case study was developed for simulation-based design, incorporating formal optimization tools and their limitations in an automated process.

A case study for application of the DSS to the design of snubbers and their locations in a piping system was conducted. The genetic algorithms are being used to not only arrive at the most optimal solution but also evaluate the design trade-off. The Modeling to Generate Alternatives (MGA) technique would then be used to evaluate alternative designs that are near optimal but different.

More significant progress than was expected was made in the development of a decision support system. In addition, its application was illustrated by a case study. This illustration was undertaken for two reasons: (1) In the opinion of the researchers, a DSS forms a key element of the process envisioned for DPCIT project; (2) the work had to be reorganized since there was insufficient information for extending the work in Year 2 on categorizing and collecting piping cost data. (It was not possible to develop similar cost data for piping and concrete structures in advanced plant designs.)

Solid Modeling-"Design to Analysis" Tool: The collaborators successfully completed a preliminary version of a "design-to-analysis" tool that converts solid models of a pressurizer and its piping systems in a computer-aided drawing (CAD) package into a finite element mesh ready for analysis. This finite element model, which incorporates the dynamic coupling between the pressurizer and its piping systems, can be used to optimize the design of piping supports as a significant procedure to reduce cost.

Seismic analyses of the finite element model of the pressurizer assembly will be performed with the appropriate floor time histories of seismic accelerations provided by Framatome ANP DE&S. The piping design details of anchor locations and constraint conditions will also be provided by Framatome ANP DE&S and incorporated into the finite element model (see figure 2).

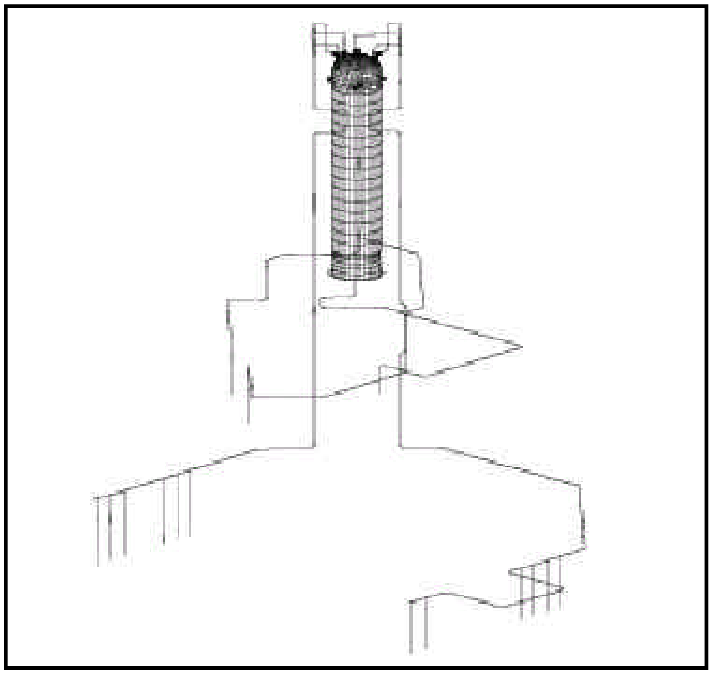


Figure 2. Coupled Model of Pressurizer Assembly and Piping Systems

4D Visualization Modeling: The team identified strategies used to reduce costs associated with the Design, Procurement, Construction, Installation and/or Testing phases of the Korean Standard Nuclear Plant (KSNP) Project, using "4D visualization." Construction data from the KSNP project was collected and the schedule reduction method used was reviewed. A KSNP 4D visualization model was developed and the design model was linked with the construction schedule for the facility's entire power block (i.e., Containment, Auxiliary, and Turbine Building).

Additionally, 4D simulation movie clips were prepared for each building and for the whole power block (see figure 3).

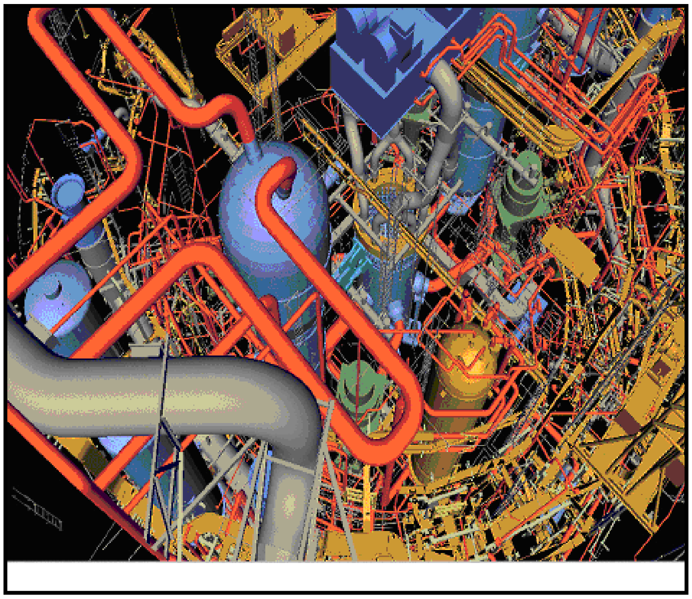


Figure 3. Inside of KSNP Containment Building

Capital Cost Model Development and Testing: The objective of this effort, conducted during the first and second year of the project, was to develop a capital costing model that addressed the uncertainty in the potential savings in both costs and project duration, separate from other research activities within this project. A tool was sought for ranking various options in terms of their ability to reduce capital costs, and for developing a methodology for converting uncertainty to risk. In FY 2001, the methodology was tested using a simplistic example. The process used a work breakdown structure (WBS) to build a factored estimate and schedule. Impacts and range of the factors and schedule durations were assessed using multiple attribute utility analysis. These were all combined in a Monte Carlo analysis to generate a probabilistic distribution of final costs and durations. Ranking of options is achieved by assessing the impact on cost and schedule by individually removing a proposed savings scheme from the model.

The final report included the development of a factored costing model and expansion of the project understanding of the impact of external constraints on project duration and final cost.

Project Management Cost and Schedule Modeling: Initial versions of two advisory systems for project management have been successfully developed: 1) A System Dynamics model that uses the deterministic approach and 2) a Bayesian Belief Network (BBN) model that uses the probabilistic approach. These two systems have been benchmarked with real-case information and showed good agreement between system predictions and real case data. They also incorporate one of the very important issues in the real project management area that has not been included explicitly in the decision-making process: the long-term benefit (or penalty) of management actions to a project. Additional results derived by the developed advisory systems were benchmarked. These results were compared to real case data or to the opinions of experts, to obtain the real case data. In addition, the "debugging" of the developed interface programs was continued, and more tests performed with the advisory systems (see figure 4).

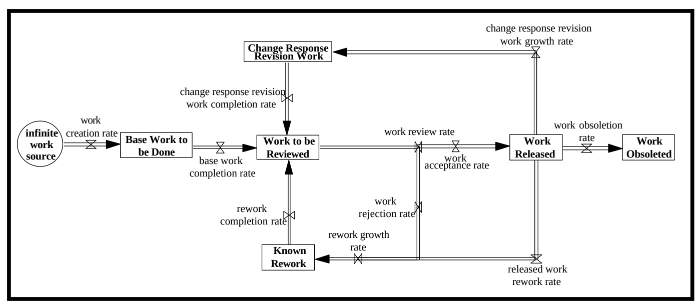



Figure 4. Work flow diagram within a particular stage of the project

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Innovative Chemithermal Techniques for Verifying Hydrocarbon Integrity in Nuclear Safety Materials

Primary Investigator: L. Mason, Pacific-Sierra Research Corporation

Collaborators: University of Virginia; University of Maryland

Project Number: 99-094

Project Start Date: August 1999

Project End Date: August 2002

### Research Objectives

This research and development program is designed to explore new methods of assessing the current condition and predicting the remaining life of critical hydrocarbon materials in nuclear power plant environments. Of these materials, Class "1E" safety cable insulation is the primary focus. Additionally, materials for o-rings, seals, and lubricant products designed for nuclear applications will also be studied. This three-phase applied research program is providing industry with new, innovative methods and reference data to conduct pragmatic programs to monitor the condition of materials for a wide range of polymer-based products. Key research objectives include development of a material-condition-monitoring database, optimization and standardization of various testing procedures, implementation of proven engineering development methodologies, and analyses of inter-technology correlations.

Milestones are defined by a five-task, work-breakdown schedule. Task 1 encompasses conducting front-end R&D program activities and planning additional tasks. Task 2 concerns identifying and reporting on subject cable materials, and their acquisition, accelerated aging, and testing by a suite of chemithermal methods. Tasks 3 and 4 comprise similar objectives for, respectively, o-rings, seals, and lubricant materials. Task 5 concerns required programmatic documentation. Under Phase 1 of this project, research progress to date fell mainly within Tasks 1, 2, and 5.

### Research Progress

The greatest fraction of Phase 1 resources for this project was expended during conduct of Task 1, the "Research and Development Plan." The next largest fraction of resources was applied to achieve progress on the first research task (Task 2), "Polymer Cable Insulation," specifically described below. In Task 1, personnel requirements were determined and fulfilled; and a new laboratory space was commissioned. Spare parts and routine supplies were acquired for three chemithermal analyzers: a Perkin-Elmer TGA7 (thermogravimetric analyzer); and two Perkin-Elmer DSC7s (differential-scanning calorimeter) instruments, one with an available high-pressure (up to 600 psig) cell accessory. Computer hardware and software upgrades included Perkin-Elmer software upgrades and an operating platform upgrade to Windows NT. These intense activities significantly expanded the laboratory's capability to simultaneously operate the suite of analyzers available at that time, as well as future additions, which now include a Perkin-Elmer TMA7 (thermomechanical analyzer) and a Perkin-Elmer Fourier transform infrared (FTIR) analyzer, the latter being configured to routinely analyze thermogravimetric analysis (TGA) combustion gases. Development of three research task plans proceeded as expected, and were focused, respectively, upon the subject materials: 1E electric cable insulation materials, o-ring and seal materials, and lubricants. Activities for each task included the following: identification of critical materials, acquisition, sample preparation, accelerated aging to simulate normal aging over 40 years under combined radiation and thermal stress conditions, chemithermal measurements, and analysis and reduction of data.

In the conduct of Task 2, accelerated aging protocols for cables were developed in the context of a rectangular matrix approach, called the "qualification aging matrix (QAM)," and were patterned after a good deal of successful research conducted previously. Twelve condition points in the QAM correspond to material conditions ranging from unaged to fully aged caused by multiple stresses throughout a full normal life in-situ. In a fully aged condition, operational integrity of these materials is expected to be intact throughout a postulated loss of coolant accident (LOCA). Plans were to analyze all condition points for a given material by chemithermal analytical methods. These methods include oxidation induction time (OIT), which measures product stability at high temperature and remaining antioxidant content (hence, remaining life); oxidation-induction temperature (OITP), which is a more rapid measure of stability and structural changes, related to OIT; and TGA, which measures thermal stability and gives some compositional information. Analytical methods for OIT and OITP tests had previously been standardized by Veridian PSR through funded government research. Standardization of these procedures involved multi-parametric experiments involving popular cable materials then under consideration. Sample mass, particle size, experimental conditions, and analyzer operating parameters and programs were all systematically investigated.

Similar experiments were conducted in this research to optimize TGA methodology. Critical cable-insulation products were identified through extensive contacts with material suppliers and users. Identification of critical cables for research is an on-going challenge, since installed-product operating data is continually updated, the number of manufacturers declines, virgin supplies of popular cables dwindle, and new products used for selective plant cable replacements become increasingly important. An array of ethylene-propylene rubber (EPR) and cross-linked polyethylene (XLPE) insulated cable products was selected, requested, and gradually amassed for this program. As these materials were being acquired, accelerated thermal aging procedures were being conducted, and chemithermal measurements of unaged and thermally aged samples were made. When enough materials had been thermally aged, gamma-irradiations were contracted for at the University of Maryland. To date, measurements of OIT, OITP, and TGA have been in line with expectations, but with some notable new discoveries, one of which is mentioned below.

The bulk of Phase 2 efforts focused on the application of chemithermal assay methods for new organic materials for o-rings and lubricants. This was the first work of its kind with these new materials. The o-ring materials studied were Nitrile-70, Nitrile-75, Butyl-70, and Ethylene Propylene-75. The lubricant materials were Krytox GPL-107 and Fomblin YR1800. OIT and OITP measurements were conducted on both aged and unaged materials in an experimental matrix that emulated the previous polymer cable measurements. The following observations were noted:

(1) The experimental aging and chemithermal assay matrix first engineered for the polymer cable materials proved to be a viable means of conducting degradation studies on other organic materials.

(2) OIT and OITP measurements conducted on these materials demonstrated the feasibility of using innovative new methods to characterize the oxidative degradation for o-rings and lubricants.

(3) TGA methods, while good for the o-rings, were not applicable to lubricants because of the material properties.

All o-ring and lubricant measurements have now been added to the comprehensive chemithermal assay measurement database.

This was successfully completed in June 2002 with the following major results:

- Establishment of a material-condition-measurement database consisting of over 1,100 OIT, OITP, and TGA measurements on 20 different types of cables, o-rings, seals, and lubricants
- A proven methodology for using OIT and OITP measurements on diverse materials that converges best practices from multiple research efforts
- A correlation study that shows the potential efficacy of correlating the following pairs of chemithermal measurement technologies: OIT-to-OITP; OIT-to-TGA; and OITP-to-TGA

The detailed results of this study are contained in the 132-page technical report entitled, Innovative Chemithermal Techniques for Verifying Hydrocarbon Integrity in Nuclear Safety Materials-Final Report (August 2002).

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Modular and Full Size Simplified Boiling Water Reactor Design with Fully Passive Safety Systems

Primary Investigator: Mamoru Ishii, Purdue University

Collaborators: Brookhaven National Laboratory

Project Number: 99-097

Project Start Date: August 1999

Project End Date: January 2003

## Research Objective

The primary goal of this research project is the scientific design of a compact modular 200 MWe and a full size 1200 MWe, simplified boiling water reactors (SBWR). Specific objectives of this research are to:

- Perform scientific designs of the core neutronics and core thermal-hydraulics for small capacity and full size simplified boiling water reactors;
- Develop passive safety system design;
- Improve and validate safety analysis code;
- Demonstrate experimentally and analytically all design functions of safety systems for design basis accident (DBA); and
- Develop the final scientific design of both SBWR systems, SBWR-200 and SBWR-1200.

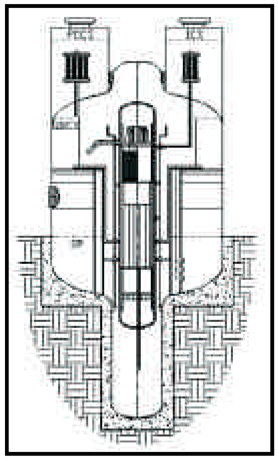


Figure 1. The schematic depicts SBWR-200 reactor containment.

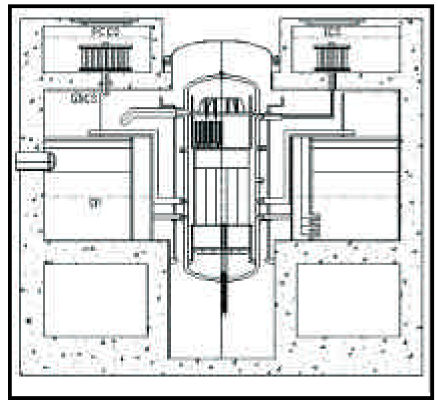


Figure 2. The schematic depicts SBWR-1200 reactor containment.

## Research Progress

The following activities have been accomplished.

- Designs were developed for the compact SBWR-200 and SBWR-1200 thermal-hydraulic system and neutronic systems. The designs are shown in Figures 1 and 2, respectively, for SBWR-200 and SBWR-1200 reactors. Development of the designs involved identification of the principal design criteria dictated by the safe operation of the reactor, identification of coolant requirements, design of the engineered safety systems, and design of emergency-cooling systems based on passive systems and scaling analyses.
- A novel passive design of the hydraulic vacuum breaker check valve (HBVC) was developed and evaluated through RELAP5 simulation. This new check valve is based on the hydrostatic head, and has no moving components. It is comprised of only one additional tank, and one set of piping to the SP and DW. This system is simple in design and hence, easy to maintain and qualify for operation. The RELAP5 simulations were performed for the SBWR-200 and SBWR-1200, with the old mechanical and new HVBC valve, for the main steam line break (MSLB) transient. The new valve performance agreed quite well with that of the mechanical vacuum breaker check (MVBC) valve.
- The HVBC valve was designed, constructed, and installed for testing in the PUMA facility. Integral tests were carried out on the PUMA with the new HVBC valve for the MSLB and bottom drain line break (BDLB) accidents. The test data for the HVBC valve were compared with the data from MVBC valve, and the agreement was very good.
- A safety analysis for an anticipated transient with scram was performed for the SBWR-1200 using RELAP5. The transient considered was a main steam isolation valve closure accident with scram. The

simulation showed that the reactor is shut down without emergency water injection, and the decay heat is adequately removed by the ICS.

- Design basis accident scenarios were studied for the safety assessment of the SBWR-1200. Large break and small break loss of coolant accident (LOCA) integral tests for the SBWR-1200 were carried out in a PUMA integral test facility. These integral tests were perfomed to assess the safety systems and the response of the emergency core cooling systems to a LOCA.
- RELAP5/MOD3 best estimate reactor thermal hydraulic code was used to model the PUMA MSLB and BDLB integral tests. The analysis was used to demonstrate the safety features of the modular SBWR design and to validate the code applicability in the facility scope. Overall, the code gave a reasonably accurate prediction of the system thermal hydraulic behaviors. This allowed an accurate assessment of the design feature of SBWR-200 and SBWR-1200 safety components. It also indicated some code deficiency that should be improved for a better simulation.
- A detailed analysis and core design was performed for SBWR-200 and SBWR-1200. The neutronics work was performed in order to (1) acquire and validate the computer codes required for the neutronics design and analysis of the SBWR (HELIOS, PARCS, and RELAP5/TRAC); (2) develop neutronics and thermal-hydraulics models of the SBWR-600 and compare the results to the RAMONA-4B predictions; and to (3) perform designs of the SBWR-200 and SBWR-1200. Core depletion calculations were performed with PARCS, for a full fuel cycle analysis. A fuel lattice design was developed to optimize the fuel cycle safety parameters. A detailed study was carried out to improve the neutronics/thermal-hydraulics of all of the SBWR models. A fuel cycle analysis of the SBWR-200 and SBWR-1200 was also carried out.
- A stability study of the SBWR-600 (GE design) SBWR-200 and SBWR-1200 under normal startup and abnormal startup has been performed. Neither the geysering instability nor the loop type instability was predicted for SBWR-200 and SBWR-1200 by RAMONA-4B in the startup simulation following the recommended procedure by GE. The density wave oscillation was not observed at all because the power level used in the simulation was not high enough. A study was made of the potential instability by imposing an unrealistically high power ramp, with pressure restricted to 1.9 bar, as suggested by GE. Core flow oscillations of small amplitude were predicted by RAMONA-4B with a period of between 31.8 and 46.7 seconds, similar to that of the TRACG prediction by GE.

## Planned Activities

The NERI project has been completed. Future plans include testing the performance of the final scientific design of the compact modular SBWR-200 and full size SBWR-1200.

NUCLEAR ENERGY RESEARCH INITIATIVE

# A New Paradigm for Automatic Development of Highly Reliable Control Architectures for Future Nuclear Plants

Primary Investigator: Richard Wood, Oak Ridge National Laboratory

Collaborators: North Carolina State University; University of Tennessee

Project Number: 99-119

Project Start Date: August 1999

Project End Date: September 2002

## Research Objective

This research focuses on development of methods for automated generation of control systems that can be traced directly to design requirements for the life of the plant. The final goal is to "capture" the design requirements inside a "control engine" during the design phase. This control engine is not only capable of automatically designing the initial implementation of the control system, but it also can confirm that the original design requirements are still met during the life of the plant as conditions change. Thus, the control implementation approach can provide a self-maintenance capability.

The control engine captures the high-level requirements and stress factors that the control system must survive (e.g., a list of transients, or a requirement to withstand a single failure). Therefore, the control engine is able to generate automatically the control system algorithms and parameters that optimize a design goal and satisfy all requirements. As conditions change during the life of the plant (e.g., component degradation, or subsystem failures) the control engine automatically "flags" that a requirement is not satisfied, and it can even suggest a modified configuration that would satisfy it.

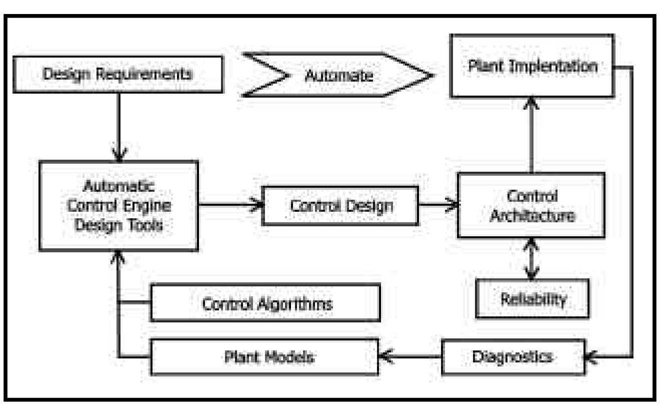



Figure 1. The schematic diagram shows the automated control design process.

This control engine concept is shown schematically in Figure 1.

## Research Progress

Project accomplishments include:

<u>Advanced Control Tools and Methods:</u> Research in this area is directed toward developing and demonstrating the control engine concept. Libraries of control algorithms have been developed, with a selected set having been demonstrated for prototypical control engine problems. The automatic control engine has been successfully applied to the generation of controllers for U-tube steam generators (UTSGs) and feedwater systems using a full plant simulation of a pressurized water reactor (PWR). In addition, the self-maintenance capability of the control engine concept has been demonstrated using fault injection with the simulated plant. In the application, a multi-step process was used. First, diagnostic modules identify a sensor failure. Next, the control system simulation model is updated to reflect current plant status. Subsequently, the control engine automatically validates the control design against the system requirements and constraints and, for this application, generates a new control solution. Finally, the new controller software is implemented and permits continued normal operation in spite of the degraded plant conditions. As additional research under this area, general methodologies have been developed for control priority mode selection and for handling the sensor and actuator nonlinearities as piecewise linear functions.

<u>Advanced Monitoring and Diagnostics:</u> The objective of this task is to develop an on-line monitoring system for fault detection and isolation of sensors and field devices in a nuclear power plant. Data-driven models have been developed for the characterization of sub-system dynamics for prediction of state variables, control functions, and

expected control actions. The fault detection and isolation (FDI) modules combine system operational knowledge (including system simulation) and a rule-based logic (with options for other fault pattern classification techniques) for identification of both single and dual faults in dissimilar sensor and field devices. The FDI techniques have been successfully applied to a laboratory process control loop and a UTSG using the full-plant PWR simulator. Demonstration of the FDI module for both single and simultaneous dual faults has been accomplished and includes the following highlights:

- Rule-based decision making
- Fault isolation using fault residuals and pattern classification
- Steady state and transient plant operation conditions
- Combination of sensors and valve actuators
- Demonstration of the functional features of the FDI module using the PICASSO interface system from the Halden Reactor Project

Nuclear Power Simulation and Reliability Methods: Research in this area involves development and application of a full plant engineering simulation code to represent the dynamic response of PWRs during normal operational transients as well as design basis events. Work under this task required the addition of a full balance-of-plant model, as well as other improvements, to the plant simulator. Options have been added to the code to allow for fault injection such as degradation in the heat transfer across the steam generator from both fouling and blocked or plugged tubes, as well as the corruption of sensor outputs through step and ramp changes in sensor output with arbitrary levels of random noise. This simulator serves as the demonstration platform for the control and diagnostic methods developed in this project. Additionally, as an element of the control system design approach, a strategy and methods have been defined to integrate and automate instrumentation and control (I&C) system reliability analysis with nuclear power plant simulation. To realize this capability, the control system simulation is structured as a reliability block diagram model representing all units that compose an I&C system. For each unit, input data consists of the type, tag number, power sources, applicable failure modes (high, low, on, of, open, closed, etc.), failure rates, linkage to all of the unit input sources, and linkage to all of the unit output destinations. Using this simulation capability, a systematic evaluation of the effect of each failure mode can be accomplished and a comprehensive failure modes and effects analysis established. A prototypic application of the basic elements for this approach was performed using representative feedwater control system architectures.

Nuclear Information System Architecture and Integration: Research addressing the control and information system architecture for future nuclear power plants involves the evolution of the Plant-Control Computing Environment (PCCE) concept. Functional requirements for the PCCE have been develop to address general design attributes, human-system interface requirements, control application interface requirements, computing platform interface requirements, monitoring and control requirements, fault handling and recovery requirements, system management requirements, and configuration requirements. A prototypical PCCE has been developed to investigate implementation issues and to serve as the framework for an integrated demonstration of the research products for this project. The PCCE provides a distributed computing environment supporting a high-level supervisory control and monitoring system (see Figure 2). Application programming interface elements have been developed to

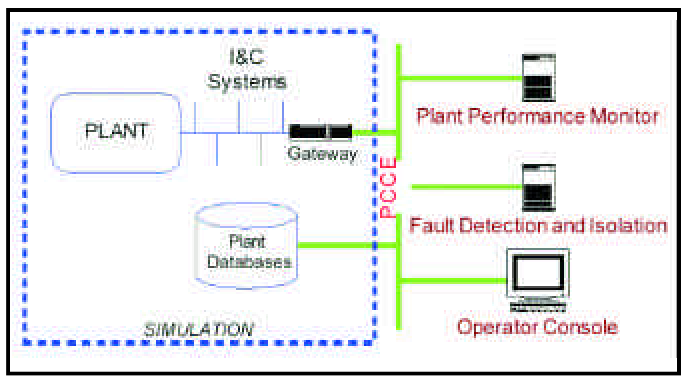



Figure 2. The graphic illustrates the plant-control computing environment and applications.

provide communication and information services for the other research modules (e.g., Fault Detection and Isolation, Plant Performance Monitor, PICASSO-based Graphical User Interface, Full-plant PWR simulator). An integrated demonstration of the research products from this project was performed using the PCCE as the interface platform. The prototypic application illustrates the interaction among the control, diagnostic, simulation, and interface elements. In addition, the self-maintenance capability of the control system architecture was demonstrated through injection of faults into the plant simulation, detection of degraded conditions by FDI modules, adaptation of the control system simulation model to reflect the current plant status, automatic assessment of the control system, and regeneration of the control solution to accommodate changes in the plant condition.

## Planned Activities

In Phases 3, an integrated demonstration of the research products from this project will be accomplished. The requirements-driven control system design will be investigated further through additional applications of the automatic control engine concept. On-line diagnostics system requirements will be determined through stand-alone and integrated (i.e., coupled control and diagnostics) implementation of the FDI modules. The automated reliability analysis methods will be developed through an application to a feedwater control system architecture.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Multi-Application Small Light Water Reactor (MASLWR)

**Primary Investigator:** S. Michael Modro, Idaho National Engineering and Environmental Laboratory

**Collaborators:** NEXANT; Oregon State University (OSU)

**Project Number:** 99-129

**Project Start Date:** October 1999

**Project End Date:** December 2002

## Research Objectives

The primary project objectives are to develop the conceptual design for a safe and economic plant and to test the design feasibility. A small, natural-circulation, light water reactor is proposed with the primary goal of producing electric power, but including the flexibility to be used in process heat applications with deployment in a variety of locations. Economic and engineering analyses will be used to address the design and safety attributes of the concept. These analyses will be coupled with testing in an integral test facility to demonstrate the concept's technical feasibility.

## Research Progress

Three major efforts were addressed in the first year of the project:

- To establish the requirements and design criteria applicable to the MASLWR
- To develop a baseline design concept, including a preliminary cost-estimate
- To generate the general scaling methodology needed to construct an experimental facility with which to test significant features of the baseline concept

The initial concept, explored during Year 1 activities, was a natural-circulation design to be operated at approximately 1,000 MWt and 5.4 MPa steam pressure. This design included four horizontal U-tube type steam generators located at a height of 36 meters above the thermal center of the reactor core. A cylindrical containment, 30 meters in diameter, housed the reactor and primary system and the required support systems and equipment. The preliminary estimates for this design indicated that the busbar cost would be about \$0.057/kWh. It was concluded that if the basic concept principles identified at the outset of the project were maintained (i.e., a pressurized water system with natural circulation), cost reduction could be achieved only by using smaller, simpler, factory-assembled units.

Consequently, activities in Years 2 and 3 have focused on developing a modular reactor design that consists of a self-contained reactor vessel assembly, steam generators, and containment. These modular units would be manufactured at a single centralized facility; transported by rail, road, and/or ship; and installed as a series of self-contained units.

Design optimization studies yielded a natural-circulation concept with a helical-tube steam generator, shown in Figure 1. Primary-side fluid flowed through the shell of the steam generator, and the secondary-side fluid was inside the tubes. The advantages of this system include low primary-side frictional losses, and the use of a single pressure vessel that encloses the reactor core and the steam generator. The primary pressure vessel is located within a containment that is a vertical, cylindrical vessel with elliptical heads. The primary vessel is partially submerged in liquid. The containment itself is submersed

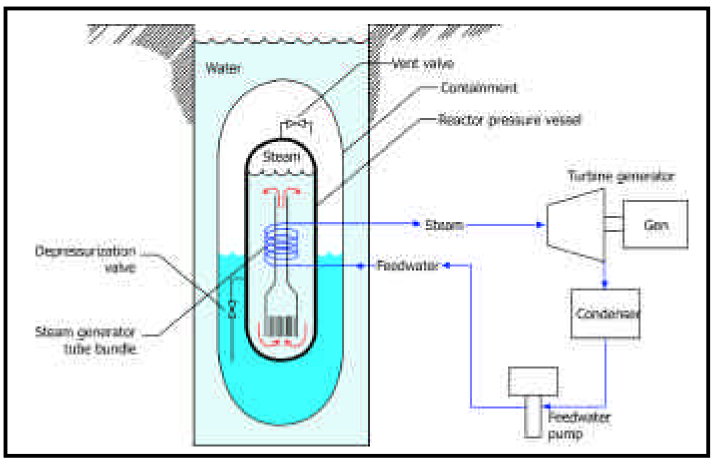



Figure 1. The schematic illustrates the natural-circulation, helical-tube steam generator of the MASLWR Baseline Design Concept.

in a below-grade pool of water, which serves as a passive ultimate heat sink.

A RELAP5 model was developed for the MASLWR to determine the operating characteristics of the design and to perform accident studies. Performance characterization studies demonstrate that the modular, natural-circulation design will operate at approximately 150 MWt and 1.5 MPa of steam pressure, which yields 35 Mwe at a thermal efficiency of 23 percent.

Safety analysis studies were performed for hypothetical accident scenarios based upon Final Safety Analysis Report Chapter 15 Guidelines for current generation pressurized water reactors, and grouped by frequency of occurrence. All the analysis cases demonstrate acceptable performance with significant margins to safety limits.

A MASLWR baseline plant consisting of 30 power generation units was used to develop schedule and cost estimates. The construction and startup schedule is estimated at 18 months for the first two power generating units, and at 36 months for the entire baseline plant. Total capital cost is estimated at $1,200/kWe and the busbar cost is $0.034/kWh.

Plant arrangement drawings have been developed for the 30-unit power generation complex and also for a six-unit plant. These drawings include the reactor and turbine generator buildings and a fuel handling and maintenance building. Other major facilities are also provided, including a main heat rejection system, control building, remote shutdown building, machine shop, and warehouse, as well as facilities for personnel services, plant services, administration and training, waste treatment, and guardhouses.

A preliminary concept was also developed for a stand-alone, single-unit plant, which would be capable of operation for a total of 60 years, consisting of two 30-year, self-reliant periods, with a core designed with a fuel cycle life of 5 to 10 years. Three to six refueling outages will be necessary during each self-reliant period. All new and spent fuel for the period will be stored onsite, and will be shipped offsite for disposal or reprocessing at the end of the period.

Seawater desalination in combination with power generation was also investigated for the 30-unit plant. The following seawater desalination processes were considered:

- Multistage flash distillation
- Multi-effect distillation
- Reverse osmosis

The results showed that reverse osmosis is the most attractive alternative. It reduces the electrical power output to 25.4 MW, and produces 15.4 MGD of desalinated water. The additional capital cost is $62.2 million and the annual power cost is $4.0 million.

District heating and/or cooling was also investigated. However, assuming a minimum practical hot water temperature of 180ºF, the turbine exhaust pressure would need to be 10 psia compared to 0.75 psia for the baseline design, thereby reducing the electric power capability from 35 MWe 18 MWe. A power reduction of this magnitude for using turbine heat exhaust for district heating and/or cooling makes the economics questionable.

A fuel handling and maintenance concept has been developed for the MASLWR. The facility will include one out-of-service module, which will be available as an immediate replacement for a module requiring refueling, thereby minimizing outage time. The modules will be transported intact to and from the refueling/maintenance facility, entirely under water. Details of the disassembly and reassembly processes have been developed. The refueling and maintenance activities are entirely automated.

The overall scaling analysis has been completed for the MASLWR test facility at OSU. It includes a Natural-Circulation Scaling Analysis, a Sump-Recirculation Scaling Analysis, a Reactor Coolant System Depressurization Scaling Analysis, and a Containment Pressurization Scaling Analysis. The analysis indicates that full power steady-state operations will be well-simulated in the test facility. The facility will obtain valuable information on helical tube steam generator performance. The power-to-helical tube-surface area scale ratio has been preserved. The unique OSU containment design preserves both the power-to-containment volume scaling and the power-to-active heat transfer surface area scaling requirements. The Automatic Depressurization System (ADS) Blowdown behavior and containment response will be well simulated in the test facility. The thick-wall vessel required to operate the facility at full pressure (1.25 in of thickness) results in too much reactor pressure vessel mass. This will likely prolong the downcomer hot-wall effect during long term sump recirculation cooling period. The hot wall effect will lead to conservative results with regards to core cooling during this period.

The GOTHIC computer code was used to determine the maximum design pressure for the test facility containment. The results were generally consistent with the RELAP5 calculations for steam vent actuation without submerged ADS operation.

Design specifications for manufacture were developed for the test facility containment vessel and its associated liquid pool based on the Containment Pressurization Scaling Analysis.

The MASLWR test facility has been completed mechanically. The containment and liquid pool vessels have been delivered and the interconnecting piping has been connected to the test loop.

Figure 2 shows a photograph of the MASLWR test facility at OSU. All of the primary loop hardware, controls, instrumentation, and related software have been purchased. Also, the primary loop of the facility and all of the supporting structure have been set in place, and

Figure 2. The primary loop and structural support of the MASLWR Test Facility have been constructed at Oregon State University.

power and controls have been installed. The instrumentation has been installed and its operation verified for reading by the Data Acquisition System. Initial measurements of the test facility's component volumes have been obtained and primary loop pressure drop measurements have been preformed. Cold Shakedown tests were performed and were repeated following replacement of a leaking hot leg riser seal.

Additional work is being performed to assess the GOTHIC condensation models and 3-D nodalization. The test facility input deck for GOTHIC has been modified to incorporate the features of the final containment design.

Programming of the instrumentation and control software for the test facility is complete. The control systems will allow OSU staff to perform testing from a separate control room. Control algorithms have been developed, and are ready for testing.

Three comprehensive papers have been peer-reviewed and prepared for presentation.

## Planned Activities

A series of Hot Shakedown tests are planned and are in progress. Phase 1 will include a series of steady state single-phase natural circulation tests to characterize the operation of the facility during single-phase conditions. The test will measure flow and power enhancement effects due to subcooled boiling, and to parameterize the performance of the helical tube steam generator as a function of feedwater flow rate and core power. The steady-state control algorithms will be be tested concurrent with the hot shakedown tests.

Phase 2 testing will consist of a several transient blowdown tests, which will include the test facility containment vessel and associated liquid pool. These tests are still being designed.

Two heater rods failed during performance of Phase 1 hot shakedown tests. The heater rod design is being analyzed to see if the two failures are related to the heater design. Current plans call for hot shakedown testing to recommence after repair of the leaky heater rod seal.

Additional work is being performed to assess the GOTHIC condensation models and 3-D nodalization. The test facility input deck for GOTHIC has been modified to incorporate the features of the final containment design.

NUCLEAR ENERGY RESEARCH INITIATIVE

# STAR: The Secure Transportable Autonomous Reactor System, Encapsulated Fission Heat-Source (ENHS Project)

Primary Investigator: Ehud Greenspan, University of California, Berkeley

Collaborators: Argonne National Laboratory (ANL); Lawrence Livermore National Laboratory (LLNL); Westinghouse Electric Company LLC

Project Number: 99-154

Project Start Date: August 1999

Project End Date: December 2002

## Research Objectives

The primary objectives of the ENHS study were to assess the feasibility of

(1) Designing lead-bismuth or lead cooled reactor cores for 20 years of full power operation without refueling and with nearly zero burnup reactivity swing, using either Pu-U or enriched uranium fuel;

(2) Designing the ENHS to have 100 percent natural circulation in both primary and intermediate coolant systems with reasonable module dimensions and weight;

(3) Designing the ENHS module to be free of mechanical connections to the energy conversion system;

(4) Designing an intermediate heat exchanger (IHX) that is an integral part of the ENHS module vessel and assessing the feasibility of manufacturing it;

(5) Fueling the ENHS module in the factory, transporting it fueled and weld sealed to the nuclear power plant site and installing it in the reactor pool;

(6) Removing the module from the reactor pool at end of life and transporting it unopened to a recycle facility;

(7) Designing the ENHS to have autonomous load-following capability;

(8) Designing the ENHS reactor to have passive safety so that postulated accidents will not damage reactor components;

(9) Attaining a high energy-conversion efficiency;

(10) Assessing the feasibility of using alternative coolants while retaining the unique characteristics of the ENHS; and

(11) Assessing the ENHS reactor ability to meet the goals set for Generation-IV reactors.

## Research Progress

The design domain has been identified for cores that can operate at 125 MWth for 20 years with nearly zero burnup reactivity swing. The core design variables include the core height, lattice pitch-to-diameter ratio, and fissile fuel contents. It was determined that it is possible to design such cores using either Pu-U fuel having approximately 11-12 weight percent Pu or uranium enriched to approximately 13 weight percent, both in alloy with Zr (10 weight percent). The core life is limited by radiation damage to the clad. A fission gas plenum volume that is equal to the fuel volume is required to accommodate the fission gas pressure buildup and maintain the clad integrity. The core designs are simple: they have uniform composition, no blanket or reflector assemblies, and a single central safety assembly. Six axially movable absorber assemblies surrounding the core are used for reactivity control. They are lifted almost completely above the fuel level to bring the core to full power. A combination of tungsten and $B_4C$ is used for the absorbing material; its specific density is larger than that of Pb-Bi so that scramming can be done by gravity. The maximum change in $k_{eff}$ throughout 20 years of full power operation is only 0.2 percent or about $0.5. After the reactor is brought to full power it may be necessary to adjust the peripheral absorber elevation approximately once every two years. The core power shape and reactivity coefficients stay exceptionally constant throughout the life-cycle. Figure 1 illustrates the extent of radial power shape variation in 20 years.

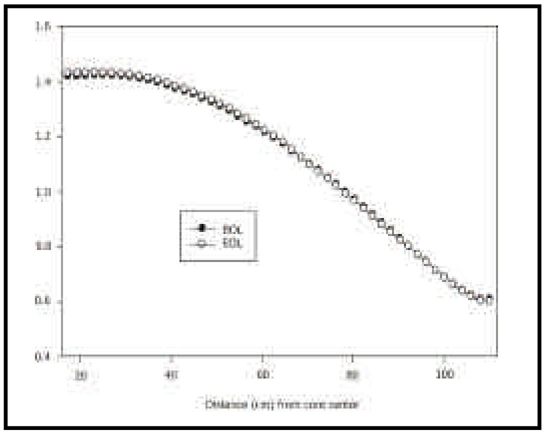


Figure 1. The graph shows axially integrated radial power distribution variation over 20 years of full power operation of the ENHS module.

It was found feasible to design ENHS modules to deliver 125 MWth from the primary to the secondary coolant through a 4-mm-thick, intermediate heat exchanger with a maximum primary coolant to maximum secondary coolant temperature drop of 50°C and with 100 percent natural circulation. The required reactor vessel height is close to 20 meters. In fact, good synergism was found between the requirement for 100 percent natural circulation and the requirement for a primary-to-secondary temperature drop of 50°C. It was also found that the ENHS reactor can maintain 100 percent natural circulation over the entire range from nominal power to decay heat power level.

A significantly more compact and lower weight ENHS module has been conceived and partially analyzed. It uses cover-gas lift-pumps to enhance coolant circulation. The gas circulators are located outside of the reactor vessel. With use of a lift pump, the ENHS module is approximately 10 meters long—about half the length and volume of the original module, with 100 percent natural circulation. The power level of an ENHS module with a lift pump could be approximately 190 MWth.

An IHX that is integrated within the ENHS module vessel walls and has practically identical heat transfer characteristics to those of the originally proposed confinement wall has been conceived and analyzed. It consists of rectangular channels circumscribing the primary coolant riser in the space between the inner and outer structural walls. This integrated IHX was found able to withstand the loads that are expected during transportation, installation, and operation. Relative to circular tube IHX, the rectangular channel IHX features close to an order-of-magnitude smaller number of channels and smaller friction losses due to elimination of grid spacers. Another innovative IHX design has been conceived; it consists of nested tube bundles. Advantages of the proposed nested channels IHX concept are that: it can be made from off-the-shelf components; it has high rigidity against buckling; and it is easier to fabricate.

A strategy was developed for fueling the ENHS module in a factory and transporting it to the nuclear power plant site as a weld-sealed unit. To avoid fuel damage during transport and module installation, the fuel is embedded in solidified Pb-Bi that fills the vessel to above the fuel rods. The total weight of the reference ENHS module for transportation is estimated to be about 300 tons. This is approximately half the weight of steam generators for large pressurized water reactors (PWRs) that have been transported from the factory to nuclear power plant sites. Dimensions of the reference ENHS module are somewhat smaller than dimensions of a large PWR steam generator. It appears feasible to factory manufacture and fuel the ENHS module and ship it to the site using available transportation equipment.

At the site, the ENHS module will be inserted into the pre-heated secondary Pb-Bi pool vessel. Hot Pb-Bi is then pumped into the ENHS vessel through a pipe. This hot Pb-Bi, along with the hot Pb-Bi in the pool, will melt the solid Pb-Bi at the lower part of the vessel. It has been found feasible to melt the primary coolant and bring it to the reactor startup temperature of 350°C within approximately two days of its insertion to the pool without having to use special external heaters in the module.

After 20 effective full power years, the ENHS module will be removed from the reactor pool into a storage vault on site. Before removal, forced cooling will be applied and the Pb-Bi will be pumped out the ENHS vessel until it reaches the upper level of the fuel rods. The rest of the Pb-Bi will be replaced by Pb. The Pb will be solidified approximately 10 days after shutdown by cooling the outer walls of the vessel and the surface of the shutdown assembly channel located at the center of the core. If cooling is provided only at the outer boundary, the Pb can be solidified after ~22 days. The ENHS module will stay in the storage vault until the decay heat drops to a level such that passive cooling will permit the Pb to remain solid during transportation. The ENHS with the solidified Pb will then be prepared as a licensed shipping package and transported to a recycling facility. Two packaging configurations were conceptually designed for shipping the used ENHS module. The first uses a cask to contain the ENHS module and is based on conventional Type B spent fuel shipping casks. The second configuration uses the ENHS module as the containment system and is based on the decommissioned Trojan and Shippingport reactor packages.

Due to its negative temperature feedback and to cooling based on natural circulation, the ENHS reactor has a load-following capability over a wide range of power. The reactor power level will adjust itself to the power demand without operator intervention. This autonomous load following capability has been demonstrated in numerical simulation. The operators need only perform a startup and a periodic surveillance function. Both functions may potentially be accomplished remotely from a centralized facility that services many units.

A preliminary safety analysis was performed for the reference ENHS design. The accidents considered so far include a startup accident, a loss of heat sink accident, and a steam line break without scram accident. It was found that under all accident conditions considered, fuel and clad temperatures will remain significantly below the safety limits and the integrity of all systems will be maintained. Figure 2 shows that in a loss of heat sink without scram accident-the worst plausible accident identified-the peak fuel temperature will not exceed its steady state operating conditions. The decay heat is passively removed by the reactor vessel air-cooling system (RVACS). Contributing to the exceptional safety features of the ENHS are the lack of pumps and valves in the primary and intermediate cooling systems, use of full natural circulation, high heat capacity, low power density, more than a 1,000°C margin between the coolant operating and boiling temperatures, and availability of very

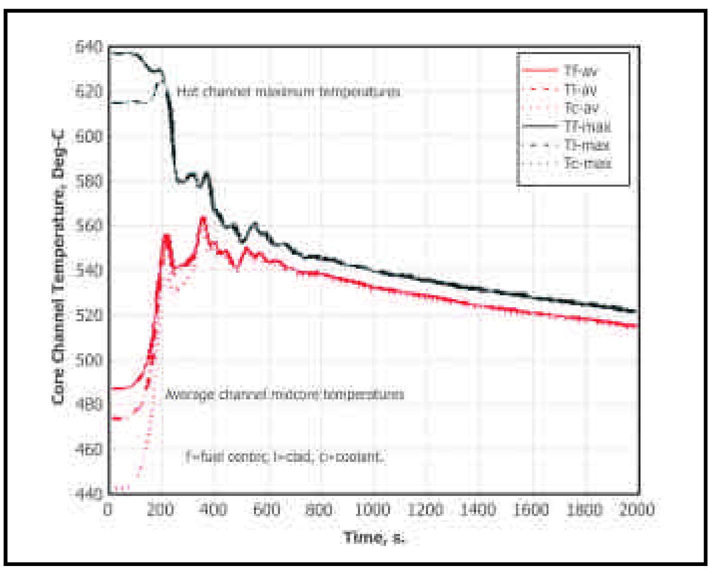



Figure 2. The plots show temperature evolution in selected core locations following a loss of heat sink accident without scram.

small excess reactivity for positive reactivity insertion accidents.

Steam generators have been designed to meet several unique requirements that are dictated by the ENHS reactor layout:

- Effective utilization of the pool volume surrounding the ENHS module
- Minimum friction losses so as to enable 100 percent natural circulation of the intermediate coolant
- Absence of a mechanical connection with the ENHS module
- Minimum flow rate of water into the intermediate coolant pool in case of a breach in steam generator tube or failure of other water-containing component
- Accommodation of a large thermal expansion
- Ease of inspection and maintenance
- Modular design that is easy to install and replace

Two-dimensional flow simulation showed that at nominal operating conditions only 3 percent of the intermediate coolant flow bypasses the steam generators. This small bypass has a negligible effect on the ENHS energy balance. The 2-D flow simulation also revealed that there is a stagnation zone at the bottom of the reactor pool. A design modification that eliminates the stagnation zone has been worked out; it involves insertion of a vertical partition in the reactor pool. Another novel plate type steam generator has recently been conceived that may allow making the reactor pool more compact or integrating the steam generator with the IHX.

A Rankine steam cycle was designed to match the heat source characteristics of the ENHS steam generators. The gross thermodynamic efficiency for converting the ENHS heat to electricity was calculated to be 40 percent; the corresponding net efficiency is 38 percent. These efficiency values correspond to a simple energy conversion system that has no reheat and moderate steam pressure of 120 bars. The main effect of the reheat is a reduction of the problematic low-pressure steam moisture. Only marginal power addition is achieved—less than 1 percent. Increasing the steam pressure to 160 bars can increase the net efficiency to 40 percent. An even higher efficiency can be attained using supercritical steam. However, the steam moisture level increases with steam pressure increase, unless reheaters are used. An energy conversion system using $CO_2$ working fluid appears to make a better match with the ENHS than a steam driven energy conversion system. The ENHS reactor can be designed to provide, in addition to electricity, process heat for desalination, for district heating as well as for industrial applications.

The neutronic and thermal hydraulic characteristics of Pb-Bi and Pb are very similar. The same ENHS reactor design can, in principle, operate with either one of those coolants. ENHS can also be designed to provide nearly zero burnup reactivity swing and 100 percent natural circulation using sodium for the primary coolant and possibly also for the secondary coolant. The sodium-cooled designs feature more compact smaller diameter cores but larger core peak to average power ratio and smaller thermal margins.

The study found that the ENHS reactor concept can meet very well all of the goals set for Generation IV reactors that have been examined, including the following:

(1) Highly sustainable energy supply. The ENHS maintains its fissile fuel inventory constant; all that is needed for reusing the fuel discharged from one module is to remove fission products, add depleted uranium and re-fabricate fuel rods.

(2) Low waste. The TRU can be recycled many times.

(3) Extremely high level of proliferation resistance. There is no access to the fuel in the host country and no access to neutrons. The host country gets energy security with no need to invest in fuel cycle technologies.

(4) Superb safety and reliability. The reasons for these features were discussed in the previous page. As a consequence of these features there is no need for emergency planning zone outside of ENHS nuclear power plant fence.

(5) Very low risk to capital. This is due to relatively low cost per module, standard design with factory assembly line fabrication, short construction time, superb safety, and large tolerance to human errors. However, the economic viability of the ENHS has not yet been examined since it was out of the scope of work. Nevertheless, general considerations and very preliminary examination of the ENHS fuel cycle cost indicate that there is a good probability that the ENHS will be economically viable despite its small unit size.

Therefore, it is recommended that the ENHS reactor concept be considered as a candidate for Generation IV reactors. A follow-on study to assess the economic viability of the ENHS reactor is suggested. Before embarking upon such an assessment, the recommendation is made to

- Define the logistics and infrastructure required for fabricating, fueling, transporting, operating and handling of the ENHS module;
- Estimate the market potential for ENHS reactors;
- Assess the feasibility of a number of design variants for the ENHS along with the maximum power and maximum discharge burnup the ENHS module can handle; and
- Work out optimized designs with greater design detail for subsystems of the ENHS reactor.

The ENHS is targeted primarily for a market of small turnkey nuclear power plants with full fuel cycle services. The potential market size for the ENHS was recently roughly estimated to be between 200 to 1,400 modules per year over the next 40 years.

More detailed description of the ENHS reactor concept and its feasibility study is documented in 35 publications.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# On-line, Intelligent, Self-Diagnostic Monitoring for Next Generation Nuclear Power Plants

Primary Investigator: Leonard J. Bond, Pacific Northwest National Laboratory (PNNL)

Project Number: 99-168

Project Start Date: September 1999

Project End Date: January 2003

## Research Objectives

The objectives of this effort are as follows:

- To focus the project on relevant Generation II, III, and IV components and reactor system aging degradation mechanisms
- To specify the experimental apparatus, configuration, and instrumentation, and to allow the selected components and degradation mechanisms to be fully exercised, measured, and analyzed
- To develop and codify a Self-Diagnostic Monitoring System (SDMS) computer architecture that is modular, allows a hierarchical diagnostic modeling approach, and is extensible to the inclusion of the full suite of degradation mechanisms; this extensibility to include all active and passive components, systems, and structures to be found in the next generation of U.S. power reactors
- To design, fabricate, and test advanced smart multi-sensor RF tag modules that will serve as the Level 3 nodes in the SDMS hierarchy; the Smart Multi-Sensor Tag (SMST) to provide for wireless data communication links between distributed intelligent sensors and the Level 2 (system level) processing nodes
- To develop the analytical methods and algorithms that will provide the diagnostic and prognostic processing to enable full condition-based operations and maintenance including first-principles life-cycle asset management; to develop a pragmatic solution needed for applications to real-world problems

## Research Progress

All of the foregoing objectives have been achieved by this project. A discussion of progress in specific topical areas follows:

Wireless Technology: Smart Multi-Sensor Tag (SMST) units were designed, fabricated, and successfully tested to show that a highly reliable fault-tolerant wireless system could be constructed. A remote, totally portable data display showing system conditions and degradation alarms was demonstrated in the laboratory.

System Modification: A heat exchanger was purchased and installed in the system along with appropriate feedback controls to maintain system temperature within acceptable limits for extended filter testing. The unit is a shell and tube two-pass stainless steel U-tube assembly that uses laboratory building water as a heat sink. Performance was found to be ± 1°C in the process stream during testing.

Analysis Methodology: The analytical approach taken by the PNNL team was completed. Called condition-based operations and maintenance (CBO&M), this method is aimed at the immediate detection and diagnosis of off-normal equipment operation and the identification of the root cause stressor(s) responsible for this condition. This approach yields a computerized real-time picture of the process problem and promises a clear understanding of the temporal nature of the solution. With such a tool, true asset management can proceed using informed decisions based on known conditions, defined degradation rates and, in most cases, accurate estimates of equipment remaining life (prognostics).

The basic concept for CBO&M stressor-based analysis centers on the fact that by understanding the stressor characteristics, an anticipatory indicator is provided for mapping subsequent damage through the activation of a resulting degradation mechanism.

The premise of this methodology is that, by not trending a performance metric per se, but by focusing on trending the stressor characteristics, a precursive relationship can be derived that will allow a much more accurate projection of the remaining useful life. Figure 1

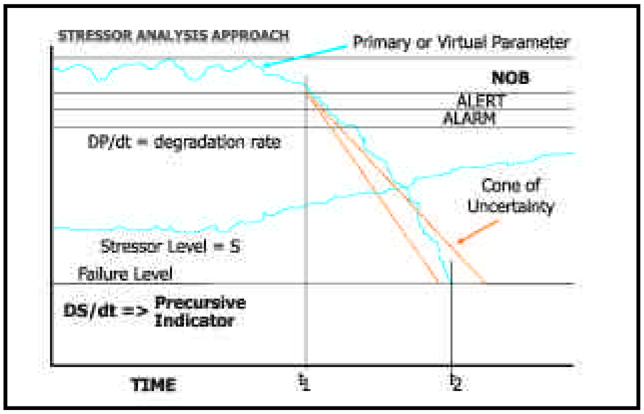



Figure 1. The graph shows the stressor measurement effect on prediction uncertainty

shows the expected result in narrowing the failure cone of uncertainty by keying on the stressor itself.

By monitoring the slope of the stressor intensity, researchers obtain a precursive operator feedback and a measure of the rate of change in the performance degradation. Thus, the stressor slope can be used to forecast and refine the path of the performance vector.

<u>Pump Testing:</u> The two most predominant mechanisms that result in centrifugal pump failure are cavitation and vibration. The instrumentation required to quantify stressor intensity and examine the physical effects of degradation have been developed for cavitation and vibration mechanisms in centrifugal pumps (Figure 2) and fouling in reverse osmosis heat exchangers (Figure 3).

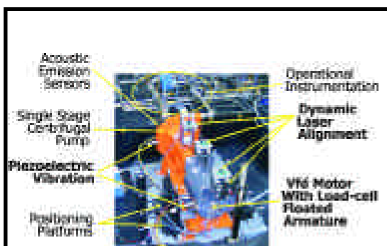



Figure 2.

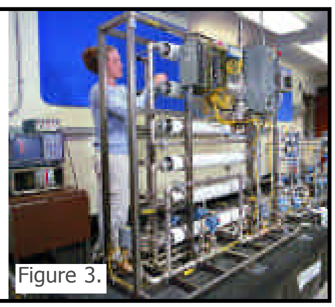


Figure 3.

Figure 2. This photograph shows the cavitation-vibration instrumentation developed for this experiment.

Figure 3. The researcher is involved in reverse osmosis testing.

<u>Vibration Mechanisms:</u> High frequency pump and motor instrumentation was developed to measure the magnitude of the stressors and resulting reaction forces on the bearings. Three measurement systems were used:

(1) Standard vibrational instruments were placed on both the pump and motor to provide a common, well-accepted reference for vibration diagnosis of machine faults.

(2) A laser dynamic position sensing system for a pump-motor set misalignment was designed and built to provide carefully quantified angular and parallel offset and to measure time-dependent response.

(3) A load cell system was developed to characterize the effects of static and dynamic rotational stressors on the armature bearings.

Analyses of the resulting spectral peaks show an excellent correspondence between the (laser) motor position indication, the vibration response, and the dynamic force loading on the bearings. Orbital and harmonic motion of the pump and motor are clearly indicated and can be readily correlated through the speactral peaks of all three sensing systems. Laser motion spectra were actually found to correlate more cleanly to the peak structure of the load cell spectra than did either accelerometer vibration sensor. By driving a three-dimensional visualization program with position data from the laser device, a clear, intuitive understanding of the primary pump-motor oscillations and their associated harmonics was obtained.

By utilizing the discrete spectral signature produced by the bearing load cells, a direct correlation between angular misalignment and the reduction in bearing life was determined. A life factor equation of the form:

$$LF = \left(\frac{P_{ei}}{P_{ea}}\right)^{p}$$

was used to derive the stressor to life reduction factor of

$$LF = 1 - (0.02) \times [\text{angular offset}]$$

where the angular offset is specified in mils of base displacement of the test pump. While not in a generally usable form since this measurement is specific to the test apparatus used, it nevertheless shows the closed form equation relating the stressor to the useful residual life (URL) of the machine. This fulfills the project goal of generating a proof of principle correlation between the primary stressor (misalignment) and the equipment URL.

<u>Cavitation Mechanisms:</u> The initial goal for the cavitation test series was to characterize the operational data as well as the spatial and spectral nature of the cavitation produced in a single stage centrifugal pump. To this end, highly accurate operational instrumentation was used to measure the motor current, suction pressure, and temperature and the discharge pressure, temperature, and flow. Specialized acoustic sensors were then installed in

the test pump per Figures 4a and 4b. These sensors were placed in direct contact with the pumped fluid to provide a clear view of the acoustic energy impacting the wall of the volute.

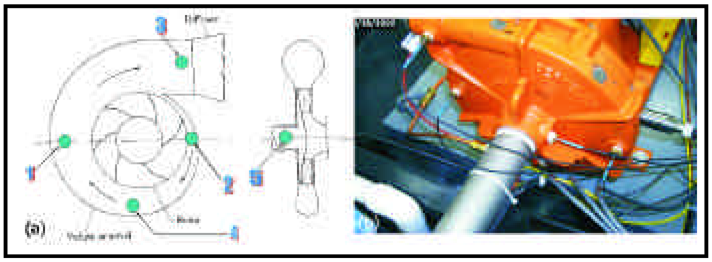


Figure 4. (a) The diagram illustrates acoustic sensor placement. (b) The photograph shows a view of the test pump.

After problems were encountered with electromagnetic interference, it was found necessary to remove the pump variable frequency drive (VFD). With the motor operating without VFD, a clear cavitation signature was obtained and signatures were obtained for both direct fluid contact and contact with the exterior surface of the pump volute. Highest values of signal-to-noise ratio were obtained using a non-intrusive probe located near the suction of the pump.

A series of tests were performed where the pump was run under conditions with varied suction pressures across the range from well above the manufacturer's suggested Net Positive Suction Head (NPSH) to the minimum suction pressure that the pump would produce. Acoustic data records were captured for each case and their respective spectra were produced using Fast Fourier Transform (FFT) processing. By normalizing the cavitation acoustic spectra to the baseline case (no cavitation), Figure 5 was the result.

Figure 5 shows the acoustic signal amplitude as a function of the suction pressure. A distinct inflection point is seen at 20 psia which has been labeled the incipient cavitation point. Above this suction pressure the literature suggests that the increasing signal is due to higher acoustic conduction through the single phase fluid. Below

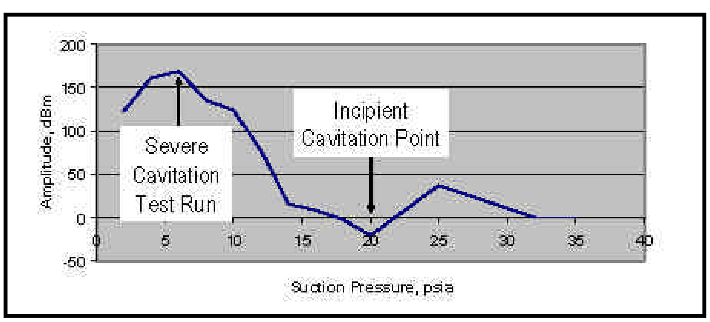



Figure 5. The graph is a cavitation test composite.

the incipient cavitation point, increased cavitation implosion is responsible for the dramatic increase in acoustic energy. It is interesting to note that the stated NPSH for the test pump equates to 18 psia, indicating the designers' lack of accuracy in determining the NPSH value, or perhaps their willingness to operate the unit under a mild cavitation condition. A maximum energy signal is seen at about 6 psia. It was speculated that these lower pressures produce a slug flow phenomena in the suction line that again reduces the acoustic signal. This hypothesis was supported by the observation of slug flow in the Lexan sections that were inserted into the suction pipe of the test rig. Figure 5 also suggests one method of scaling the intensity of the acoustic energy that results in metal removal.

A continuous cavitation run was initiated on September 2 and continued 24 hours a day for 4 weeks (with the exception of a 4-hour power outage in the laboratory). The test pump was then secured, drained, and disassembled to obtain wear readings relative to the baseline. With the exception of the wear ring clearances very little metal removal was observed. The impeller to volute gap in this area indicated a 10 mil increase in clearance.

Without performing further cavitation runs, only a simple linear correlation can be derived from the available two point data set. When combined with the acoustic intensity measurement this provides a "zeroth order" approximation to a correlation that relates suction pressure differential from incipient cavitation (the primary stressor) to the degradation rate of the pump. Making several major assumptions about the linearity of a logarithmic intensity scale and the validity of its relationship to metal removal rate, it is possible to derive an equation of the following form:

$$MRR = K\left[10\exp\left(13.9\, PSID_{NPSH}\right)\right]T(da)$$

Where:

- MRR is the metal removal rate
- T is the cavitation time in days
- $PSID_{NPSH}$ is the differential pressure between the operating point and the pump NPSH limit
- K is a material and geometric constant dependent on the specific pump
- The coefficient 13.9 is the slope of the (logarithmic) acoustic intensity line from Figure 5 and is in db/psid.

The task then remains to relate this to the useful residual life through an understanding of pump performance as a function of wear ring clearance. When the pump internal circulation reduces its throughput to below process discharge or flow requirements, the pump would be considered to have "failed."

The pump performance correlation is a second example that demonstrated that the project achieved the goal of providing a foundation for a first-principles prognostic algorithm.

Data Integration: This task centered on the completion of communications interfaces and the integration of the six (DSOM Operational Instrument Display, Dynamic Laser Alignment, Vibration Accelerometers, Bearing Dynamic Load Cell System, Acoustic Emission Array, and Ultrasonic Fouling Meter) independent technology information systems (TIS). This required upgrades to the SDMS Software architecture design and development of additional hardware and software interfaces. The goal was to integrate the TIS outputs into the Decision Support for Operations and Maintenance (DSOM) system to facilitate data transfer, diagnostics, and display (see Figure 6).

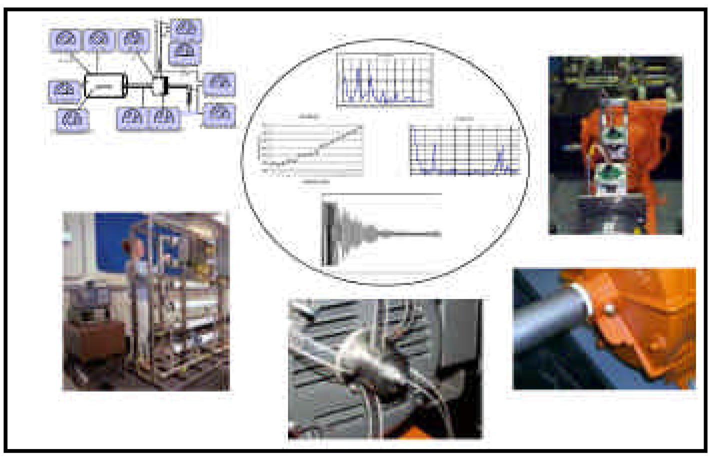


Figure 6. The graphics illustrate Multi-Technology Diagnostic Reasoning.

A software module was developed to provide a generic means to transfer spectra obtained from FFT processing from distributed processors to the DSOM system to support diagnostic analysis. Large data sets, on the order of tens of kilobytes, result from the high sample rates and the number of elemental data points required for useful frequency analysis. Transfer of such large data sets to the DSOM system would be highly inefficient particularly since the diagnostic analysis centers around only certain subsets of the data. Instead, the data sets are reduced through the distributed processing capability designed into the SDMS. The resulting data provided to the DSOM system is only that required by the diagnostic algorithms, thus reducing transmitted data by more than a factor of 100.

The DSOM diagnostic logic then selects and analyzes specific frequency bands to compare maximum peak height and integral area within the selected band with baseline data. Logic trip levels are set based on these comparisons and the result is passed to the operator via a diagnostic alert. This new integrated approach allows a prognostic evaluation to be made, and the operator can now make informed decisions based on equipment damage rates and their effect on the projected equipment residual life.

Fouling Monitor: An ultrasonic "fouling meter" was implemented on the reverse osmosis (RO) filters using a suite of 0.5 and 1.0 MHz transducers. Preliminary testing is shown in progress in Figure 3. The ultrasonic data demonstrate that pulse-echo and transmission measurements can provide a non-destructive on-line real-time monitor for filter condition assessment. Data have been shown to be sensitive to fouling layer (during both fouling and cleaning), solution concentration, membrane condition, and filter internal structure, including changes during operation. Preliminary data indicate that these can potentially be used to predict rate of change in filter condition and form the basis for the application of prognostics to the development of various forms of fouling. Similar approaches can potentially be implemented in heat exchangers and other elements in process plants where deposition or erosion is encountered.

## Planned Activities

The project has met all its major goals. Data produced by the NERI SDMS experiments have been used to generate a new and definitive set of correlations linking degradation stressors to resulting degradation rates and failure prognostics. The correlations are pragmatic in their formulation and will be applicable to current as well as future generations of fossil and nuclear reactor power plants.

The methodology developed through this project points the way to real-time diagnostic/prognostic predictions that will pay significant dividends in terms of risk reduction, root cause resolution, equipment reliability, and resource management under normal and emergency conditions. The NERI project has been completed.

# Concept Analysis of a Modular 50-MW(Th), Pebble-Bed, High-Temperature, Gas-Cooled Reactor for Process Heat

Primary Investigator: Dennis R. Liles, Los Alamos National Laboratory

Collaborators: Texas A&M University

Project Number: 99-188

Project Start Date: August 1999

Project End Date: September 2000

## Research Objectives

The overall research objective was to develop an updated concept for a small, modular, high-temperature, helium-cooled reactor that could be used to produce hydrogen by reforming methane and steam. Such an application demonstrates the use of nuclear-generated heat as an alternative to burning fossil fuels to produce process heat.

The main goal of this study was to explore one principal option for coupling nuclear reactor heat to an endothermic chemical process: specifically, a small (50-MWth), modular, pebble-bed reactor coupled to a steam-methane reforming system. Important aspects of reactor design and safety, fuel performance, and system requirements are discussed. In particular, various nuclear fuel compositions, a change in the fuel-particle coating (from SiC to ZrC), and their influence on the reactor's physics and safety are analyzed.

## Research Progress

In the high-temperature, gas-cooled reactor (HTGR) core, the use of coated particle fuel creates a double heterogeneity, both on a microscopic level (fuel particles) and on a macroscopic level (fuel element). This double heterogeneity must be considered in the neutronics calculations. Apart from a double heterogeneity, the use of helium as a coolant means that the pebble-bed HTGR core contains spaces that, because of the low density of the gas, can be considered as voids for neutronics calculations.

The major parts of detailed neutronics calculations were carried out with the SCALE and MCNP code systems. In SCALE resonance self-shielding calculations, the spatial self-shielding due to the heterogeneity is considered using Dancoff correction factors. Additional calculations were performed to determine the appropriate model for a correct description of such a complicated system as the pebble-bed core. SCALE is an extremely important tool for fuel performance evaluation because it can perform accurate fuel-depletion and fission-product-buildup calculations, thus accounting for cross-section and flux changes due to fuel irradiation. The MCNP code was used for final verification of the model.

The innovative use of composite materials such as carbon-carbon composites and zirconium as a coating for the fuel particles increases the margin of safety in high-temperature regions of the system. The proposed system is shown schematically in Figure 1; parallel components are not shown to improve clarity.

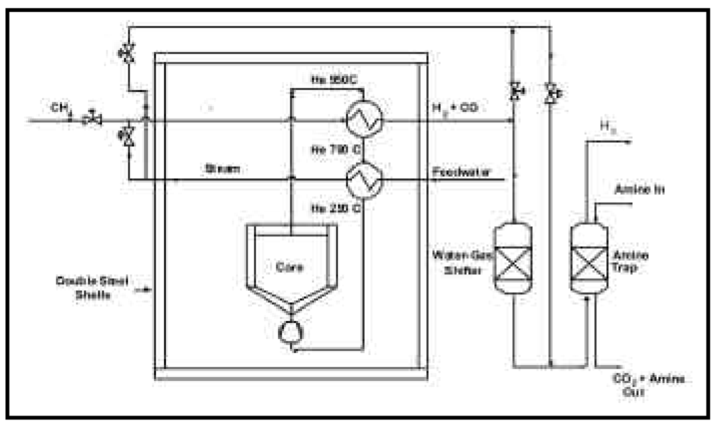



Figure 1. The schematic shows the design for a small modular HTGR for steam-methane reforming.

The primary circuit shown above consists of helium at 40 bars driven by dual circulators upward through the packed pebble bed. The helium enters at 250°C and leaves at a mixed mean temperature of 950°C. Carbon-carbon ducts connect the reactor to the first heat exchanger, the process unit. The helium enters the shell side of the heat exchanger and passes through once. Regenerative bayonet tubes contain the methane, steam, and process gases. Helium exits at 700°C and enters the steam generator, again on the shell side. U tubes contain the water-steam mixture. The helium exits at 250°C and returns to the circulators. Both the process stream and the water enter their respective heat exchangers at a pressure only a few bars over the primary system pressure to minimize stress on heat-exchanger components.

Steam-methane reforming is a catalyzed equilibrium reaction between methane ($CH_4$) and steam ($H_2O$) to produce hydrogen gas ($H_2$), carbon monoxide gas (CO), and carbon dioxide gas ($CO_2$). Additional processing can lead to either the separation of relatively pure hydrogen gas or the production of methanol. Alternatively, the product stream can be optimized to generate a mixture of carbon monoxide and hydrogen, which can be transported easily over long distances in a closed loop (so that the gas is recycled). These gases then recombine to produce methane and steam, an exothermic reaction releasing usable heat at the destination. The goal of the current project is to produce a relatively pure hydrogen gas. Steam-methane reforming is a well-established, commercial method for producing hydrogen gas in chemical processing plants. However, $CO_2$ is still a product of the process and must be considered. The reactor/reformer system proposed here reduces the $CO_2$ emission only by using a reactor to generate the process heat that eliminates burning fossil fuel. Further reduction in $CO_2$ emission requires application of advanced systems that have not yet been proven commercially.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Novel, Integrated Reactor / Power Conversion System (LMR-AMTEC Project)

Primary Investigator: Pablo R. Rubiolo, Westinghouse Electric Company LLC

Collaborators: University of New Mexico (UNM); New Mexico Institute of Mining and Technology

Project Number: 99-198

Project Start Date: August 1999

Project End Date: August 2002

## Research Objectives

The overall objectives of this project are to assess the feasibility, develop engineering solutions, and determine a range of potential applications for a Novel Integrated Reactor/Energy Conversion System. The goal is to design a power supply for use by developing countries and in remote locations that is proliferation-resistant, reliable, and economical. The main features of this project are the development of a long life liquid metal reactor (LMR) (without refueling up to 10 years), and of a static conversion subsystem comprised of an Alkali Metal Thermal-to-Electric (AMTEC) topping cycle and a Thermoelectric (TE) Bottom cycle. In addition, various options of coupling the LMR with the energy conversion subsystem are being explored.

The project is being performed by the Westinghouse Electric Company LLC, which is responsible for the long-life sodium reactor development; the University of New Mexico's Institute for Space and Nuclear Power Studies, which is developing the AMTEC/TE energy conversion system; and the Institute for Engineering Research and Applications (IERA) at New Mexico Institute of Mining and Technology, which is responsible for the design of the electric converter modules and supports Westinghouse's activities related to transport safety and waste disposal.

## Research Progress

The research progress and achievements are reported in this section by project participant.

Westinghouse Electric Company LLC

The work falls into three major areas:

Selection of the reference LMR-AMTEC design concept

Different design options were evaluated using a plant model. An Indirect Coupling (IC) plant with Alkali Metal Boilers (AMB) (see Figure 1) was chosen as the reference design, since it exhibits the best performance. The main features of the design are as follows:

- Two independent loops are employed for the IC between the LMR and the AMTEC units. No shielding is required for the secondary loop.
- Sodium and potassium are used as primary and secondary coolants, respectively.
- The net plant efficiency is 28.2 percent, when the core outlet temperature is 1,070°K.
- The LMR core is composed of 78 fuel elements (13 control rod assemblies) and 78 reflector elements. The fuel is (U,Pu)N and the cladding is made of the refractory alloy Nb-1Zr.
- The AMBs generate the potassium vapor, which is fed into the AMTEC units. The AMB can operate in once-through or in recirculation mode (with a vapor separator).
- The LMR is a pool reactor, the AMB and the primary pumps are placed inside the reactor vessel, hence excluding large LOCAs (Lost of Coolant Accident) by design.

Operating parameters of the LMR-AMTEC

The following plant parameters and components were determined and studied: working temperatures; flow rates and pressures; core design (fuel and cladding); alkali metal boiler design and operation; primary pumps characteristics; flow-induced vibrations in fuel elements and AMB tubes; corrosion allowance; reactor vessel design; and the in-vessel layout. A first economic anlysis of the plant was also performed.

Safety features of the LMR-AMTEC

The work concerning safety aspects of the LMR-AMTEC system includes the following:

- Reactivity control systems: An actively controlled absorber rod bundle assembly was adopted as a reactivity control system. Boron carbide was selected as the control rod absorber material. The studies of the reactivity behavior upon sodium removal showed an acceptable core reactivity response.
- Heat removal systems: The secondary loop, composed of the alkali metal boilers and the AMTEC units, was chosen as a normal Decay Heat Removal System (DHRS) of non-safety grade during hot and cold shutdown. In addition, a safety grade Passive Heat Removal System (PHRS) is proposed as an emergency DHRS. The PHRS of the LMR-AMTEC is activated by changes of the sodium levels that occur after the primary pumps are tripped.

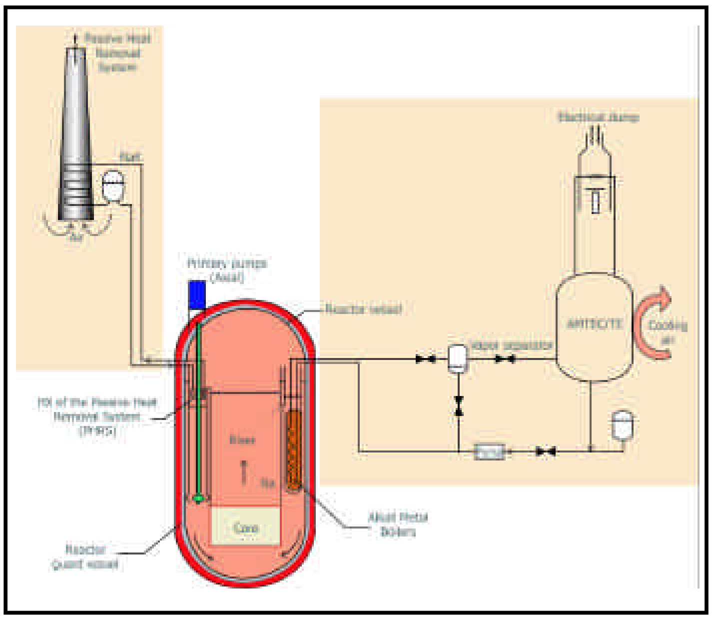


Figure 1. Schematic showing LMR-AMTEC plant design.

Institute for Space and Nuclear Power Studies (ISNPS) (University of New Mexico)

The converter, which comprises an AMTEC top cycle and a PbTe TE bottom cycle on the condenser side of the AMTEC, was designed and optimized for maximum overall conversion efficiency, for both sodium and potassium working fluids. The AMTEC topping cycle of the power conversion units delivers high power (>40 kWe) and high voltage (~400 V). The operating temperature of the evaporator (beta-alumina solid electrolyte, BASE) is approximately 1,000°K and approximately 1,121°K, for potassium and sodium working fluids, respectively. The heat rejected by the condenser of the AMTEC flows to the bottom cycle of TE modules, through a conductive coupling arrangement. The electricity generated by the TE bottom cycle, which is cooled by natural convection of ambient air, contributes between 10 percent and 30 percent of the total electric power generated by the AMTEC/TE converter units.

Performance analyses of the AMTEC/TE converter units showed that a total conversion efficiency in excess of 30 percent could be achieved, based on conservative assumptions regarding the technology of the AMTECs, and using off-the-shelf technology of lead-telluride (PbTe) TE modules. As more advances are made in the technology, higher conversion efficiencies, in excess of 34 percent for the combined AMTEC/TE converters, and a long operation lifetime of 5 to 10 years, with little degradation, would be possible in the near term.

The interfacing arrangement of the plant designs developed and investigated by the UNM-ISNPS was obtained by providing the reactor thermal power to the AMTEC/TE converter units through a heat exchanger (HX). The overall thermal and electrical performances of the plant were evaluated using a thermal-hydraulic model of the secondary loop of the LMR-AMTEC. In these designs, the secondary sodium or potassium liquid exiting the HX is partially evaporated in expanders placed in each AMTEC/TE converter. These studies showed that a Na/K plant (sodium in the primary loop and potassium in the secondary loop) could deliver a net power output of 25.4 MWe at an overall conversion efficiency of 28.6 percent, while for a Na/Na plant, the net electrical power output is 25 MWe at an overall plant efficiency of 27.7 percent. In addition, these analyses showed that the K-AMTEC/PbTe converters have an efficiency higher than that of the Na-AMTEC/PbTe converters. However, the K-AMTEC/PbTe converters deliver an electrical power output lower than the Na-AMTEC/PbTe, requiring the use of 25 percent more converter units in the Na/K plant and as a result, increasing the cost per kilowatt-hour.

Regarding the thermoelectric bottom cycle, the bottom cycle to the condenser of the top cycle was designed and thermally coupled. The use of different thermoelectric materials (instead of PbTe) for the bottom cycle was also investigated, including both single- and multisegment thermoelectric couples. Finally a performance model of the fully integrated sodium-and potassium-AMTEC/TE converters was developed. This model was used to optimize the unit's design for maximum efficiency and to investigate and determine the operation regime in which the static AMTEC/TE converters are load-following.

Additional work includes the study of different high-energy uses and the nuclear power plant options for those applications.

Institute for Engineering Research and Applications

The work performed by the IERA included topics related to transport safety, corrosion control and waste disposal of the LMR-AMTEC. Based on the selected design of the LMR-AMTEC components and the coolant types, the wastes were classified and characterized according to Code of Federal Regulation.

## Planned Activities

The NERI project has been completed.

# Nuclear Energy Research Initiative

## Direct Energy Conversion Fission Reactor

**Primary Investigator:** Gary Rochau, Sandia National Laboratories

**Collaborators:** Sandia National Laboratory; Los Alamos National Laboratory; General Atomics; University of Florida; Texas A&M University

**Project Number:** 99-199

**Project Start Date:** September 1999

**Project End Date:** December 2002

### Research Objectives

The U.S. Department of Energy, Nuclear Energy Research Initiative (NERI) Direct Energy Conversion (DEC) project began in August of 1998 with the goal of developing a direct energy conversion process suitable for commercial development. In the first two years of the project fission processes that capture the energy of the fission fragment were examined through electromagnetic or magnetohydrodynamic principles. Roughly two-thirds of the project has been completed and investigators believe that a viable direct energy device is possible. Three concepts are under conceptual development: a Fission Electric Cell using magnetic insulation, a Magnetic Collimator using magnetic fields to direct fission fragments to collectors, and a Gas Vapor Core Reactor using magnetohydrodynamics (MHD) to generate electrical current. The concept-definition effort has focused on detailed examination of the physics of each concept and on building an engineering model to examine the design options for a power plant. The original plan was to select only one concept at the end of the second phase, but it is now believed that too many details require further exploration before such a selection can be made.

### Research Progress

During the first phase of the study, nine different concepts were investigated. These concepts were analyzed and ranked to select the top three. All the selected concepts use a magnetic field in various configurations to extract energy from the fission fragments. The Quasi-Spherical Magnetically Insulated Fission Electric Cell or Fission Electric Cell (FEC), the Fission Fragment Magnetic Collimator (FFMC), and the Gaseous Vapor Core (GVC) reactor have been investigated in greater detail during Phase 2 with the objective of understanding how these concepts might be scaled to power reactor. Phase 3 efforts have focused on completing the concept definition and outlining proof of principle experiments.

<u>Fission Electric Cell:</u> This concept (Figure 1) originally used spherical cells with the fissioning cathode placed at the center. High-intensity, shaped magnetic fields trap the electrons near the cathode allowing the more massive fission fragments to reach the anode and deposit their charge.

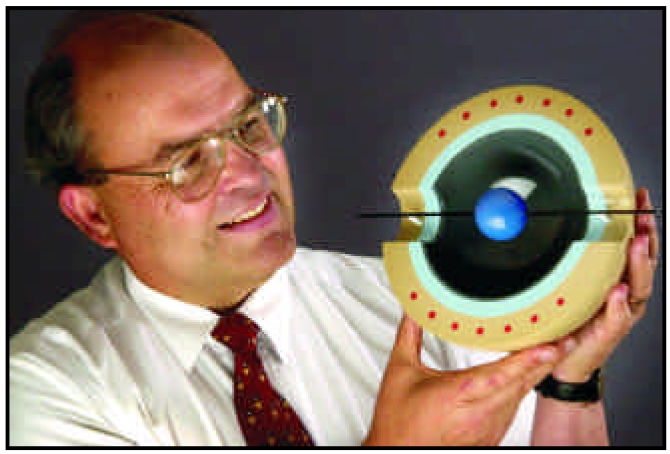


Figure 1. The individual in the photograph is holding a model of the Fission Cell under development.

Because a spherical geometry maximizes the recovery of fission particles, this scheme could theoretically achieve efficiencies as high as 60 percent. Unfortunately, this design has an extremely low fuel density, making a critical assembly very difficult. Additionally, the material generating a magnetic field absorbed too many neutrons. New configurations have been developed using externally generated magnetic fields in slab, cylindrical, and spherical geometries. Overall, these devices would be very compact but have a low power density (between 0.005 and 0.015 watts per cubic centimeter). These configurations have been modeled in terms of performance, thermodynamics, and criticality.

Issues associated with this design are the size of the assembly, ability to insulate small vacuum gaps to 20 M V/cm, cathode mechanical stability, maintenance of criticality, and conversion efficiency. These issues direct researchers to look at geometries that are more easily constructed (slab and cylindrical), trading system efficiency for mechanical stability. Low efficiencies and criticality issues are guiding them to examine on-line refueling techniques to maximize fuel burn-up and utilization. Experimental designs are being studied to establish the scientific feasibility of the FEC. These experiments examine the ability to insulate small vacuum gaps, cathode stability, formation of a secondary cathode, and fission fragment charge collection. The economic feasibility of the FEC is also being studied. Initial estimates indicate that electrical power can be generated for a cost of between \$0.02 and \$0.09. Electromagnetic modeling of the transport of electrons in the spherical geometry is being performed during the Phase 3 study. These calculations are providing insight to the transport of electrons from cell to cell and providing a basis for experimental designs.

<u>Fission Fragment Magnetic Collimator:</u> This theoretical device uses magnetic fields to direct positive and negative charges to common collectors at both ends of a solenoid magnet. Fissionable material, in thin wires or threads, is placed in a parallel magnetic field inside a cylindrical vacuum chamber. As the fuel fissions, the electrons and positive fission fragments remain separated and drift to the ends of the cylinder. At the ends, the particle energy is collected in an electric-field insulated collector.

A performance model of the FFMC has been developed incorporating an electromagnetic code to track electrons and fission fragments to the collector devices. The model indicates that overall efficiency as high as 55 percent can be achieved with thin fiber-type fuel elements. This design avoids the cathode stability issue of the FEC in that high electric fields are avoided in the region of the thin fissioning fuel. Further investigation of the charge collectors is required to establish the engineering design and to identify any stability issues in the collector area. Future experiments are being defined to focus on the degree of charge separation and possible space charge effects that may limit the power density of the system.

<u>Gaseous Vapor Core Reactor:</u> The third approach to direct conversion uses MHD to generate electricity coupled with a more conventional steam cycle to achieve high conversion efficiency. This direct scheme uses a high-temperature gaseous core reactor to generate ionized fissioning plasma that passes through an MHD channel to generate electricity. The unprocessed heat from the MHD cycle is transferred to a superheated Brayton cycle (gas turbine) and/or Rankine power cycle (steam turbine) to achieve combined efficiencies on the order of 60 to 70 percent. The concept has several advantages over existing reactor designs in that it embodies an extremely simple reactor core design with a fully integrated fuel cycle design that includes minor actinide burning. The design has a low fuel inventory: three orders of magnitude lower than conventional light water reactors. The design has disadvantages in that material temperatures are greater than 2,500°K and more research is needed in the fission-enhanced conductivity of the vapor fuel (crucial to the MHD efficiency) in addition to the immature state of MHD power conversion.

Future work will focus on material issues, conductivity of vapor fuel, and design of the purification system.

## Planned Activities

The final phase of the project is now being completed. Lacking sufficient resources for performing critical technology experiments, the team cannot make an effective selection of one concept on the basis of physical data. Consequently, the first activity in the third and final year of the project is to move toward a "Final Concept Definition" for each of the three concepts suitable for refinement and publication.

Equally important to the final project phase is the definition of experiments to provide preliminary proof of principle experiments for each of the concepts. These experiments will highlight the path forward for developing the concepts into a viable energy source. Throughout the entire process, the intent of the project has been to provide the technical basis for an alternative commercial energy source. The final report is expected to provide continuity between this effort and any future design development and commercialization efforts.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Novel Investigation of Iron Cross Sections via Spherical Shell Transmission Measurements and Particle Transport Calculations for Material Embrittlement Studies

Primary Investigator: Steven M. Grimes, Ohio University

Collaborators: University of Florida; National Institute of Standards and Technology

Project Number: 99-228

Project Start Date: August 1999

Project End Date: December 2002

## Research Objectives

The principal objective of this project is to study neutron scattering interactions in iron as a means of investigating the well-known deficiency that exists in reactor pressure vessel neutron fluence determinations. The spherical-shell transmission method is being used, employing iron shells with different thicknesses and neutron time-of-flight measurements of the scattered neutrons, to determine precisely specific energy regions over which deficiencies in the non-elastic scattering cross section for neutron scattering in iron appear to exist. Analysis of the experimental data involves correlating the data with theoretical calculations of neutron transport through the iron spheres in order to evaluate the degree to which the calculated neutron spectra predict the measured spectra relative to different types of particle interactions. In the process, new methodologies were developed for performing neutron transport calculations that are useful to a range of transport problems.

## Research Progress

Spherical shell transmission measurements were performed using neutron time-of-flight spectroscopy and accelerator-based neutron sources at selected energies greater than 1 MeV using both the $^{15}N(p,n)$ and D(d,n) sources. These measurements were conducted on two different high-purity iron spheres with shell thicknesses of approximately 3 cm and 8 cm. These two selections for sphere wall thickness were chosen in order to enhance the experiment's sensitivity to the iron cross sections, based on Monte Carlo simulations of the experiment that indicate a range of iron shell thicknesses from approximately 4 cm to 8 cm should be optimal. These measurements reveal information on the total non-elastic cross section, and various components of the non-elastic cross section for which there are neutrons in the exit channel, and provide a way of determining the quality of evaluated microscopic cross section data by an application to a macroscopic system through which neutron transport can be determined.

The larger most recently fabricated iron sphere was fabricated from high-purity iron with a low carbon content that was hot-rolled and forged to produce cylindrical billets. The iron sphere was constructed in the form of two hemispherical sections that were cut from the cylindrical billets; this fabrication work was performed using a numerical controlled lathe at Ohio University. The new sphere has some major advantages over the first sphere that was used. The new sphere was fabricated with less interior void space, which allowed the sphere to have a greater annular thickness without incurring a large increase in its outer diameter. This aspect is important relative to the alignment of beamline components and the selection of appropriate neutron beam collimation. The purity of the iron is also higher, thereby allowing for more accurate modeling of the experimental results. In addition, a new gas cell was designed to provide good charged-particle beam collimation and current integration capacity. Three concentric tubes of stainless steel constitute the major pieces. The outer tube contains a 1-mm thick gold beam stop at its end. The middle tube holds a tungsten foil approximately 5 micrometers thick at its end. The inner tube holds the final beam collimator and charge suppression. Each tube is soft-soldered to a brass cylinder that has o-ring seals. Between the inner tube's brass cylinder and the outer and middle tubes, there is a piece of Teflon to insulate the outer two tubes for charge collection.

Experimental runs were first started by collecting necessary calibration spectra to determine the efficiency of the NE-213 being employed. For this task, the Al(d,n) reaction was used, which was determined to be slightly better known than the B(d,n) reaction. A stopping aluminum target was used along with a deuteron energy of 7.44 MeV at an angle of 120 degrees for these calibrations. This energy and angle were those used for the source spectrum determination, which had been carried out previously relative to the $^{235}U(n,f)$ standard neutron cross section. Measurements of the bare source were made for the $^{15}N$ (p,n) reaction at angles of 0, 15, 45, 60, 90, 100, 120, and 135 degrees using the NE-213 detector. These measurements were made using 5.1 MeV protons emerging from the tungsten foil into the 3-cm gas cell (which was maintained at 1.5 atmospheres). A large number of angles was used for both the $^{15}N(p,n)$ and the D(d,n) source reaction work in order to provide detailed source information for the computer simulations. Additional measurements were also made with the small sphere and the large sphere for this source at angles of 0, 45, 90, 120, and 135 degrees using a NE-213 detector. A series of additional source measurements were conducted for the D(d,n) reaction using the NE-213 detector. These measurements were made at angles of 0, 15, 45, 60, 90, 100, 120, and 135 degrees for 3.0 MeV, 5.0 MeV, and 7.0 MeV deuterons emerging from the tungsten foil into the gas cell, which was maintained at 2 atmospheres. Corresponding runs were also made at zero degrees with no gas in the cell. Runs were then made at these three deuteron energies with the large sphere surrounding the source. For each deuteron energy, measurements were made at 0, 45, 90, 120, and 135 degrees. Random and differential linearity runs were performed to calibrate the time per channel.

Detailed particle transport calculations have been performed to optimize the experiment, to improve the accuracy of the experimental data, and to permit testing of neutron cross sections for comparing the measurements to calculations of the neutron transport through the shells. A series of time-dependent neutron transport calculations were first employed to investigate different experimental configurations in order to optimize the experiment. For this task, the $A^3$-MCNP (Automated Adjoint Accelerated MCNP) computer code and the three dimensional Parallel Environment Neutral-particle TRANsport (PENTRAN) code were utilized in a parallel computing environment. For this work, two PC clusters (PCPEN and PCA3MC) are in use at the University of Florida. The experimental data is being analyzed using both Monte Carlo and deterministic discrete ordinates neutron transport techniques in order to obtain information about energy regions where problems may exist with accepted iron cross section evaluations.

Additional work on modeling the neutron transport has focused on improving the accuracy of the model and speeding up the calculations. The Monte Carlo model has been revised to accommodate a more accurate representation of the geometry and materials existing in the source region and the collimator. A simplified treatment of the source gas cell has been applied, and the source has also been modified to allow for full rotation, which simulates the accelerator's beam swinger facility. The Monte Carlo model has been further updated to better incorporate the experimentally determined neutron detection efficiency, which is used to scale the scoring of the detector tally. The experimental runs are being analyzed using the revised geometry and treatment of the detector efficiency. Since the Monte Carlo simulation times for these models are excessively long, effort has been made to identify that part of the simulation that is the most time consuming. It was determined that although approximately 80 percent of the simulation time is spent tracking neutrons throughout the iron sphere due to the large number of collisions in this region, a large number of these collisions result in neutron tracks that fail to scatter into the detector. More specifically, only about 1 out of 100,000 collisions in the sphere are associated with "neutrons" that are subsequently detected. This result has then been the focus of further study resulting in the development of a suitable Monte Carlo variance reduction methodology, and it was concluded that the most effective variance reduction methodology for this problem is source biasing. In this method, the source neutrons that travel towards the detector are given more weight while the neutrons traveling backwards are weighted much less. This approach results in greater sampling of those neutrons that are more likely to contribute to the detector response. Because of parallel processing and the implementation of variance reduction techniques, a speed-up of almost two orders of magnitude has been achieved while obtaining a statistical uncertainty of about 5 percent. Preliminary results show good agreement between the experimental data and the calculated flight times for those cases where the iron sphere is not in place, thereby indicating that the model of the neutron source and time-of-flight environment are adequate. This is shown, for example, in Figure 1 for the D(d,n) reaction with 5 MeV deuterons and an angle of zero degrees.

## Planned Activities

On-going and planned activities are focused on analyzing and understanding the results obtained from those cases in which the iron sphere was in place surrounding the neutron source. In Figure 2, the same conditions are in place as for Figure 1, but with the smaller sphere in place. One comparison is shown in which the simulated spectrum follows the same trends in shape as the experimental data. However, it is clear from

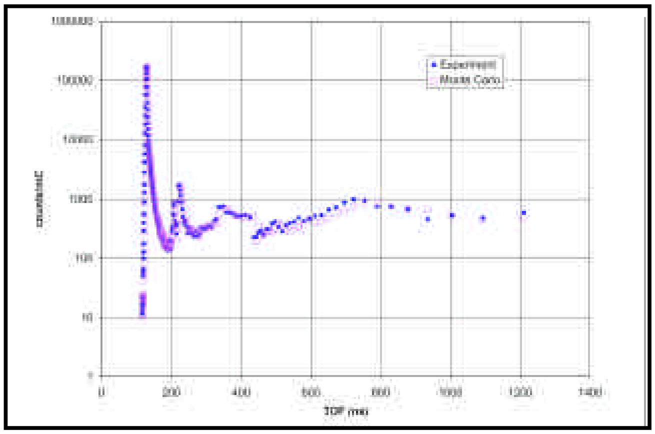



Figure 1. The graph indicates there is close agreement between the experimental and theoretical spectra when the iron sphere is not in place near the neutron source.

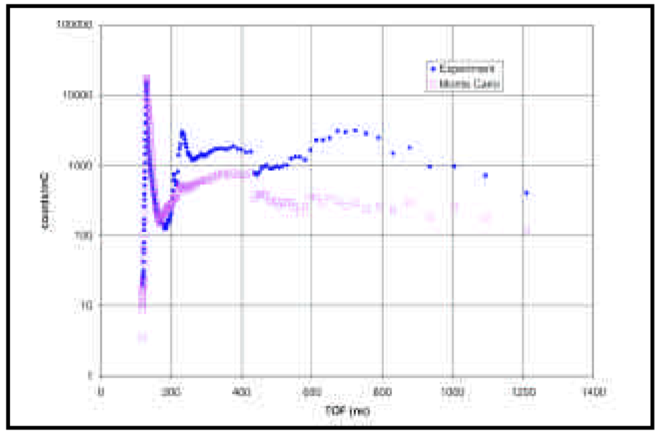



Figure 2. When the small iron sphere is in place surrounding the neutron source, there are significant differences between the experimental and theoretical spectra.

the figure that significant differences exist between the two (calculated and experimental) spectra. The process is underway to attempt to identify the source of these differences, and to determine whether they represent artifacts of the simulation process, or are indeed a manifestation of the result of deficiencies with the evaluation of the cross sections for iron.

NUCLEAR ENERGY RESEARCH INITIATIVE

# High-Efficiency Generation of Hydrogen Fuels Using Nuclear Power

Primary Investigator: Lloyd C. Brown, General Atomics

Collaborators: University of Kentucky; Sandia National Laboratories-Albuquerque

Project Number: 99-238

Project Start Date: August 1999

Project End Date: October 2002

## Research Objectives

Hydrogen is an environmentally attractive transportation fuel that has the potential to displace fossil fuels when coupled with fuel cells. Fuel cells are more efficient than conventional battery/internal combustion engine combinations and do not produce nitrogen oxides during low-temperature operation. Contemporary hydrogen production is primarily based on fossil fuels-more specifically on natural gas. When hydrogen is produced using energy derived from fossil fuels, there is little or no environmental benefit. Currently, there is no large-scale, cost-effective, environmentally attractive hydrogen production process available for commercialization.

The objective of this research is to find an economically feasible process for the production of hydrogen, by nuclear means, using an advanced high-temperature nuclear reactor as the primary energy source. Hydrogen production by thermochemical water splitting, a chemical process that accomplishes the decomposition of water into hydrogen and oxygen using only heat or, in the case of a hybrid thermochemical process, by a combination of heat and electrolysis, could meet these goals.

## Research Progress

Phase 1 of the project concentrated on finding a thermochemical cycle suitable for development into an economic process for the production of hydrogen based on thermal energy from an advanced nuclear reactor. An exhaustive literature search was performed to locate all thermochemical water-splitting cycles. Thermochemical water-splitting is the conversion of water into hydrogen and oxygen by a series of thermally driven chemical reactions. The 115 cycles located were screened using objective criteria, to determine which can benefit, in terms of efficiency and cost, from the high-temperature capabilities of advanced nuclear reactors. The 25 cycles remaining after the preliminary screening were subjected to more rigorous analysis. As part of this second stage screening process, detailed investigations were made into the viability of each cycle. The most recent papers were obtained for each cycle, thermodynamic calculations were made over a wide temperature range, and each chemical species was considered in each of its potential forms (gas, liquid, solid, and aqueous solution). As a result of this analysis, two cycles were rated far above the others: Adiabatic UT-3 and sulfur-iodine cycles. The sulfur-iodine (S-I) cycle was selected for detailed evaluation after considering the advantages and disadvantages of each cycle. Although the S-I cycle is projected to have a significantly higher efficiency than the Adiabatic UT-3 cycle, there were other reasons for selecting the S-I cycle for this work:

(1) An extensive analysis of the Adiabatic UT-3 cycle was recently completed in Japan, whereas the last complete flowsheet for the S-I cycle was developed in 1981.

(2) Major process improvements have been suggested in the literature for the S-I cycle, which have not been incorporated into the flowsheets.

(3) The UT-3 cycle requires major materials development if it is to operate in the proposed mode.

A very simplified schematic flow diagram of the sulfur-iodine cycle is shown in Figure 1.

The diagram indicates standard state thermodynamics of the chemical reactions at the indicated temperatures but much of the work on the process involves the non-ideality of the real thermodynamics. In fact, the key to the process is that the extreme non-ideality of the system separates the hydrogen iodide from the sulfuric acid in the presence of an excess of iodine.

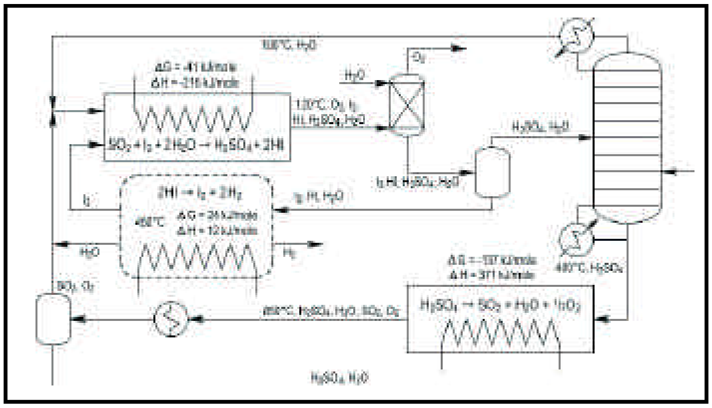


Figure 1. The schematic is a simple flow diagram of the sulfur-iodine thermochemical cycle for potentially economical production of hydrogen for transportation fuel cells.

The major undertaking during the last two years of the project was the definition of a chemical process, based on the S-I cycle, and coupled to an advanced nuclear reactor. The end result will be a flowsheet for the process, an equipment list with sizing information, and a preliminary cost estimate for the production of hydrogen from the reactor/chemical plant complex.

The last complete flowsheet for the S-I cycle was developed in 1981. This was in the early days of chemical process simulation and neither the computer algorithms nor the chemical thermodynamic models available at that time were capable of describing the very non-ideal chemistry of the cycle. Although attempts were made to use computer simulation to describe the sulfuric acid portion of the process, the flowsheet was ultimately developed using hand calculations based primarily on individual experimental data points.

Even now, after the development of chemical process simulation codes, convergence is quite difficult for very non-ideal chemistry. The lack of suitable thermodynamic models also remains a major impediment to process development. Non-ideal thermodynamic models are now being developed for common systems, but it was necessary for researchers to develop a thermodynamic model for sulfuric acid since the range of operating conditions encountered in the S-I process is far different from that encountered in normal chemical processing. Most sulfuric acid processing is performed at low temperatures. Thermodynamic models were available for sulfuric acid at low concentrations at moderate temperatures, and at high concentrations at low temperatures. However, no model was available at the high concentrations and temperatures of the S-I process.

An electrolytic non-random, two-liquid (ELECNRTL) model of sulfuric acid was developed by regressing the available thermodynamic data. Much of the pertinent data was not widely available and had not been used in previous attempts to model sulfuric acid. Aspen Plus was then used to model the sulfuric acid concentration and decomposition steps of the process. An upper bound on the hydrogen production efficiency of 61 percent was obtained by assuming that no heat would be required for the other process steps. Any heat required for the HI processing and hydrogen production step will decrease the efficiency of this value.

The system $HI/I_2/H_2O$ is even more problematic. Data was readily available for the pure components and for the two-component system, $HI/H_2O$, but there was a paucity of other data for the system. However, the mutual solubilities of $I_2$ and $H_2O$ were available along with a few liquid-liquid equilibria measurement for the high HI two phase region and some gross vapor pressure measurements of three component mixtures. The gross vapor pressure measurements were compromised by the equilibrium decomposition of HI into $H_2$ and $I_2$. Nevertheless, it was possible to regress the data to obtain an ELECNRTL model for the system. Figure 2 shows a phase diagram for the system $HI/I_2/H_2O$ calculated from this model.

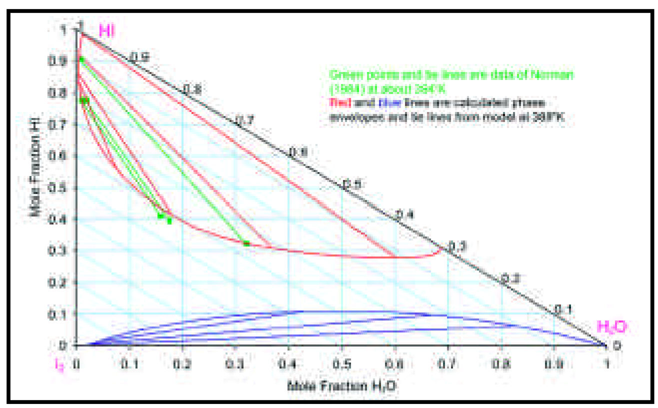



Figure 2. The figure illustrates a $HI$-$I_2$-$H_2O$ ternary diagram at about 390°K.

Two alternative versions of the $HI/I_2/H_2O$ portions of the flowsheet are being investigated. The first is a modification of the process proposed in the original flowsheets, which avoided the complexity of the $HI/I_2/H_2O$ equilibrium by extracting water away from the $HI/I_2$ with concentrated phosphoric acid. This process step was efficient but very capital-intensive. Vapor recompression was used to separate the water from the phosphoric acid. The use of co-generation of electricity and hydrogen production was investigated for increasing the overall efficiency of the combined operation while decreasing the capital cost. Electricity can be produced at 51 percent efficiency using a Brayton cycle and 850°C helium from a high-temperature nuclear reactor nuclear. If it was assumed that that the waste heat from the Brayton cycle was used to separate the water from the phosphoric acid, the overall co-generation process efficiency reached 52 percent. Unfortunately, under this scenario, most of the energy goes into electricity production and only 2 percent of the total plant output is hydrogen. This is a very acceptable efficiency for a niche market but the ratio of hydrogen to electricity is too low to support a hydrogen economy. The ratio of hydrogen to electricity can be increased at the expense of the overall efficiency. At an overall efficiency of 50 percent, hydrogen represents 7 percent of the energy output and at 47 percent efficiency—the efficiency of the original hydrogen process—hydrogen is 14 percent of the output. At this level, a significant fraction of the fuels for surface transportation could be provided by hydrogen from nuclear reactors, if all of the Nation's electricity was produced from the same reactors.

The second $HI/I_2/H_2O$ process option employs reactive distillation of the three-component mixture to produce the hydrogen. This process option should have a significantly reduced capital cost compared with the other option but it has not yet been possible to converge the model.

Meanwhile, procedures for estimating the capital cost of the process plant have been adapted to the materials and unit operations of the S-I process. The procedures have been exercised with an early version of the flowsheet and are ready for use when convergence of the latest version of the flowsheet is completed.

Sandia's nuclear engineers evaluated alternative nuclear reactor concepts to select a reactor concept to be matched to the S-I process. Beginning with a list of nine reactor types, the most promising configuration of each was selected and evaluated with respect to its potential for powering the thermochemical cycle. The results are provided in Table 1.

It was determined that the reactor heat source should not in itself present any significant issues related to design, safety, operation, or economics. Pressurized and boiling water reactors, organic-cooled reactors, and gas-core reactors were found to be unsuitable for the intended application. Although alkali metal-cooled and liquid-core reactors are potential candidates, they present a significant development risk for the intended application. Heavy metal-cooled reactors and molten salt-cooled reactors show promise in being capable of meeting the requirements. However, the cost and time required for their development are uncertain and may be appreciable. Gas-cooled reactors have been successfully operated in the required 900°C coolant temperature range, and do not present any obvious design, safety, operational, or economic issues. The study concluded that a Gas-cooled reactor, employing helium as the coolant, be matched to the S-I process.

**Table 1. Evaluation of Nine Reactor Types for Use with the S-I Process.**

| Reactor Type | Recommendation |
|---|---|
| 1. Pressurized Water Reactors | Unsuitable (Insufficient temperature) |
| 2. Boiling Water Reactors | Unsuitable (Insufficient temperature) |
| 3. Organic-Cooled Reactors | Unsuitable (Insufficient temperature) |
| 4. Alkali Liquid Metal-Cooled Reactors (lithium-cooled) | Potential |
| 5. Heavy Liquid Metal-Cooled Reactors (lead-bismuth cooled) | Promising |
| 6. Gas-Cooled Reactors (helium cooled) | Recommended |
| 7. Molten Salt-Cooled Reactors ($2LiF-BeF_2$ cooled) | Promising |
| 8. Liquid-Core Reactors (Molten Salt-Core) | Potential |
| 9. Gas-Core Reactors | Unsuitable (Unacceptable development risk) |

## Planned Activities

The third phase of the project is almost complete. The remaining scheduled tasks are the completion of the $HI/I_2/H_2O$ portion of the flowsheet, and the generation of a cost estimate for the production of hydrogen using nuclear energy.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Smart Equipment and Systems to Improve Reliability and Safety in Future Nuclear Power Plant Operations (Smart-NPP)

Primary Investigator: Felicia A. Durán, Sandia National Laboratories

Collaborators: Framatome ANP; Korea Power Engineering Company; Pennsylvania State University; Massachusetts Institute of Technology; Westinghouse Nuclear Automation

Project Number: 99-306

Project Start Date: August 1999

Project End Date: December 2002

## Research Objective

The goal of this research is to design, develop, and evaluate an integrated set of tools and methodologies that can improve the reliability and safety of advanced nuclear power plants (NPPs) through the introduction of smart equipment and predictive maintenance technology. This will ultimately aide in the reduction of construction, maintenance, and operational costs.

To accomplish the goal, the following activities were carried out under the Smart Equipment project:

- Identified and prioritized NPP equipment that would most likely benefit from adding smart features.
- Developed a methodology for systematically monitoring the health of individual pieces of equipment implemented with smart features (i.e., "smart" equipment).
- Developed a methodology to provide plant operators with real-time information through smart equipment Human-Machine Interface (HMI) to support their decision making.
- Demonstrated the methodology on a selected component.
- Expanded the concept to system and plant levels that allow communication and integration of data among smart equipment.

For this project, smart equipment embodies elemental components (e.g., sensor, data transmission devices, computer hardware and software, HMI devices) that continuously monitor the state of health of the equipment in terms of failure modes and remaining useful life, in order to predict degradation and potential failure and inform end-users of the need for maintenance or system-level operational adjustments.

## Research Progress

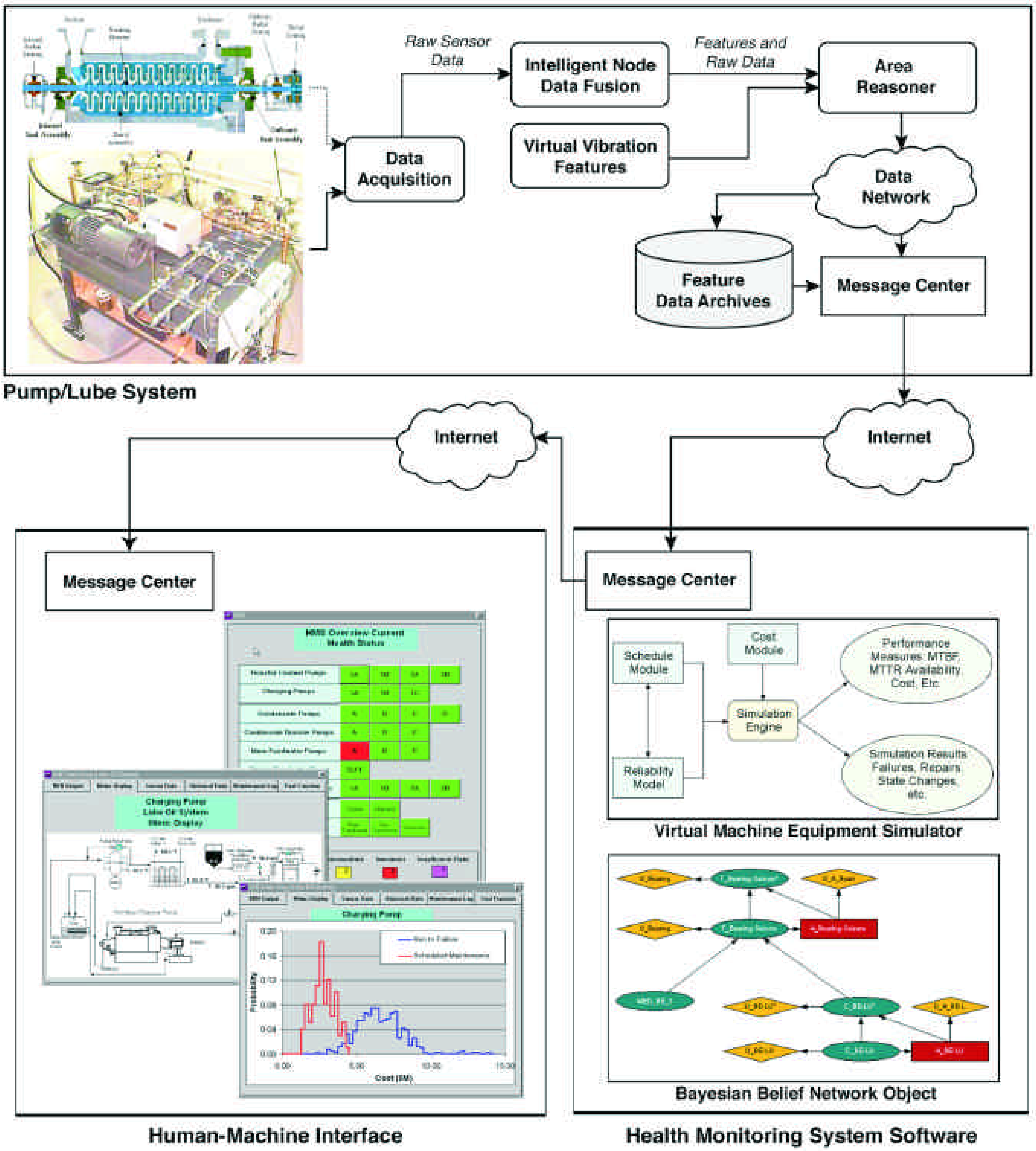
During the course of this project, the Smart-NPP team has accomplished the following:

- Completed a system prioritization study for smart equipment applications, prioritized both pressurized water reactor (PWR) and boiling water reactor (BWR) systems, and selected a high-energy, horizontal centrifugal pump as a demonstration component for a Health Monitoring System (HMS).
- Developed an HMS architecture, using Bayesian Belief Networks (BBNs) and completed detailed BBNs to relate sensor features to fault conditions for the charging pump and its lube system.
- Completed a Beta Version of the Virtual Machine Equipment Simulator (VMES) to provide a capability to simulate equipment behavior over future time intervals in order to evaluate the overall benefits to system performance from designing in smart features as well as the consequences of alternative maintenance options. Data collected on PWR equipment performance from 1990-1995 was analyzed to test the VMES and to illustrate its capability for simulating equipment failures and maintenance. The purpose of this example was to test the reliability simulation capability of VMES and to provide a limited validation of its simulation algorithms. Additionally, a sample scenario was evaluated to illustrate the use of VMES in evaluating alternative scenarios that could be considered in response to an HMS notification of a

pending equipment failure. The basis for selecting the preferred scenario will be the cost of electricity not generated as a result of a scheduled or forced outage.

- Reviewed and assessed sensor technology and instrumented the lube system test bed with sensors, including a PC104 smart sensor, that provide real-world data over the Internet to the HMS.
- Reviewed and assessed HMI technology and then developed and demonstrated an HMI as part of the integrated system.
- Instrumented the test-bed with sensors, including a PC104 smart sensor, and will provide real-world data over the Internet to the HMS.

Figure 1. Integrated Smart Equipment Demonstration Health Monitoring System Architecture



- Implemented the architecture and messaging format for the HMS to accommodate remote communication and integration among the elements of the systems.
- Established a collaborative agreement with the Korea Power Electric Company (KOPEC) for a parallel KOPEC project to develop an HMS for a control rod drive mechanism (CRDM).
- Procured the use of a demonstration test-bed of the pump lube system built at Pennsylvania State University, as the physical real world system.

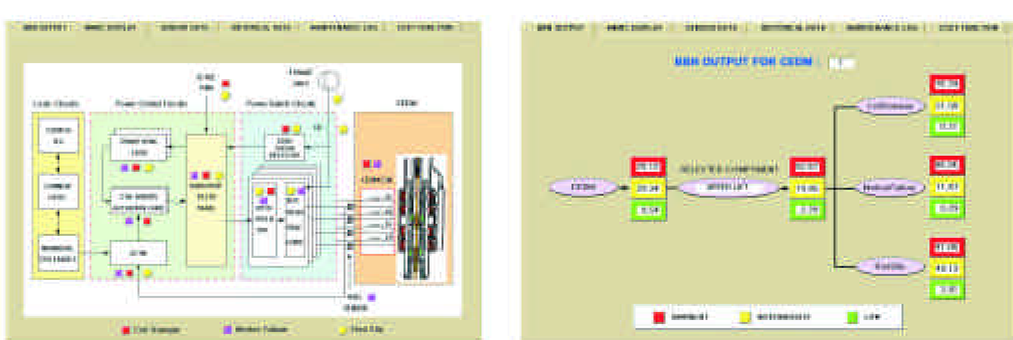
In the final phase of the project, the Smart-NPP team completed project activities to achieve the project goals. Final demonstration versions of each element of the HMS were completed, demonstration scenarios were developed to exercise the system, and the Smart-NPP team presented an integrated demonstration HMS (Figure 1) for a high-energy, horizontal, centrifugal charging pump. The demonstration HMS can replicate faults for a physical, real-world system as well as implement other faults "virtually"; process sensor features and relate these to system fault conditions; generate consequences of alternative maintenance options; and provide indications and details of system health and maintenance options on an HMI.

Additionally, collaboration with KOPEC continued, and the KOPEC team completed a successful application of the Smart-NPP methodologies to develop a demonstration HMS for the CRDM (Figure 2).

## Planned Activities

The NERI project has been completed.

Figure 2. KOPEC Smart Equipment System for a Rod Control System.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Continuous-Wave Radar to Detect Defects within Heat Exchanger and Steam Generator Tubes

Primary Investigator: Gary E. Rochau, Sandia National Laboratories (SNL)

Collaborators: New Mexico State University (NMSU)

Project Number: 99-308

Project Start Date: September 1999

Project End Date: January 2003

## Research Objectives

The goal is to design, fabricate, and demonstrate a radar system that will be translated internally through the steam generator tubing of nuclear power plants to find defects within the tube walls. The primary technical objective of this tool is the detection of incipient cracks that are about 20 percent, or less, of the tube-wall thickness. This will be about a two-fold improvement over present eddy-current technology. The second goal is to provide 100 percent volumetric examination of the tube wall at a translation speed of 40 inches/second. Thirdly, pattern-recognition algorithms will be developed to classify and size defects.

## Research Progress

The research on this project is performed in four parallel tracks:

(1) Mechanical design of the probe system,

(2) Computational modeling of the electromagnetic performance of the probe,

(3) Laboratory prototyping of the probe system, and

(4) Electronic data collection.

The mechanical design of the probe and the test fixture was assigned as a problem for senior students in a capstone, mechanical engineering senior design class at NMSU. An early effort was to build a scaled-up version of the instrument and measure its performance in a large metal tube, and to then build a smaller version. The two different sized instruments and three types of material for their fabrication were examined, both structurally and electro-magnetically, and a 6-foot length of Inconel 600 was obtained with an ID of 3.438 inches and a 0.1-inch wall thickness. The test fixture has been fabricated, and a commercial probe translation system has been integrated into the fixture. The design of the scaled-up prototype probe, together with its centering system, is complete (see Figure 1). Prototype units for the transmitter power amplifier as well as the ferrite rod antenna have been designed and fabricated. On the receiver side, a monopole antenna and amplifier have been designed and a prototype unit fabricated. Data communication circuits have been designed, built, and tested to transmit command and control data to the probe and collect data from the probe via fiber optics. Mechanical systems for driving the probe have also been designed and implemented to complement a Z-tech commercial drive system.

The project was described at the Sixth Balance-of-Plant Heat Exchanger NDE Symposium in June 2000, to solicit industrial inputs. U.S. Patent No. 6,271,670, "A Radar Back-Scatter Probe to Detect External Cracks from within a Metal Tube," was issued on 7 August 2001 and assigned to SNL. In the same month, a paper on "In-Tube Radar" was presented by researchers from both SNL and NMSU to the International Conference on Structural

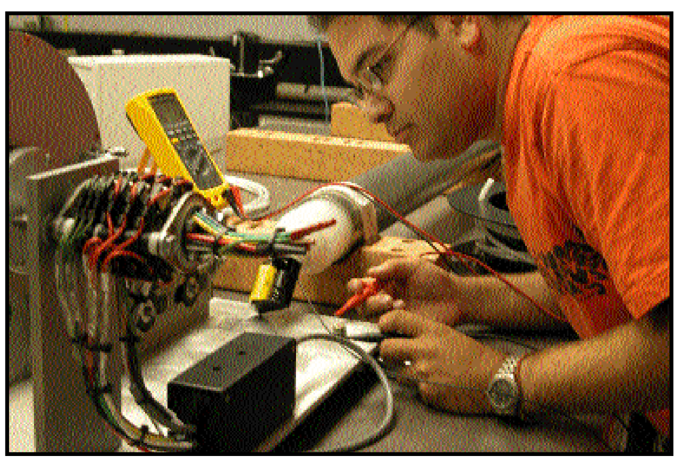


Figure 1. A scientist works on the In-tube Radar Large Scale Experiment to detect cracked tubing in nuclear power plant steam generators.

Mechanics in Reactor Technology (SmiRT 16) in Washington, D.C.

The SNL code, CTUBE, which is used to compute the internal distribution of the electric field, now provides results for unsymmetrical conditions so that it is possible to make calculations for an ITR that is tilted within a steam generator tube. CTUBE has been used to predict the cross talk voltages that will be induced upon the monopole detector. As a result, the computation of both the internal and backscattered fields that are necessary to predict Signal-to-Cross talk ratios have been done at SNL. While acceptable signals can be calculated, the intensity is extremely sensitive to alignment and requires high sensitivity equipment to detect defects in a small tube. Consequently, very specialized detector construction is required.

The laboratory design of the probe has proceeded according to schedule. The electronic and electromagnetic design of the probe, using the best technology available, continues to evolve to improve signal quality and reduce the noise observed in testing. The probe design is extremely sensitive to shielding and this has become a focus of the design revisions. The requisite design for the probe may exceed the current capability of the developers. To eliminate noise and spurious signals, an electromagnetically sealed system with ultra-precision circuitry may be required.

The project has had significant student involvement at New Mexico State University. Four Master's degree students have been supported by the project (one in Electrical Engineering and three in Mechanical Engineering). Three senior engineering capstone design teams have worked on various aspects of the project including probe design, mock up prototype design, and electrical design, and a total of 24 undergraduate students (in Electrical Engineering, Mechanical Engineering, Industrial Engineering, and Technical Writing) have been partially supported by the grant and have made significant contributions towards the advancement of the project.

## Planned Activities

A demonstration of the scaled probe, using the best available technology, is planned for December 2002.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Study of Cost Effective Large Advanced Pressurized Water reactors that Employ Passive Safety Features

Primary Investigator: James W. Winters, Westinghouse Electric Company LLC

Project Number: 00-023

Project Start Date: August 2000

Project End Date: September 2003

### Research Objectives

On December 16, 1999, the U.S. Nuclear Regulatory Commission (NRC) issued Design Certification of the AP600 standard nuclear reactor design. This culminated an eight-year review of the AP600's design, safety analysis, and probabilistic risk assessment. The AP600 is a 600-MWe reactor that utilizes passive safety features which, once actuated, depend only on natural forces such as gravity and natural circulation to perform all required safety functions. These passive safety systems result in increased plant safety and have also significantly simplified plant systems and equipment, resulting in easier plant operation and maintenance. The AP600 meets NRC's deterministic safety criteria and probabilistic risk criteria with large margins.

The large safety margins of the AP600 can be attributed to the performance of the passive safety systems in response to accidents. An extensive AP600 test program was performed to provide confidence in the ability to adequately predict the performance characteristics of the passive safety systems as required by 10 CFR 50. This test program consisted of separate effects and integral systems tests of the passive safety systems and is well-documented in NUREG-1512, Final Safety Evaluation Report Related to Certification of the AP600 Standard Design. Westinghouse used the test programs to develop analytical computer codes that can predict with adequate certainty, the performance of the passive safety systems in response to design basis and beyond design basis accidents. In addition to the extensive test program conducted by Westinghouse, the NRC also performed confirmatory tests and analyses at both the APEX test facility at Oregon State University and the ROSA test facility at the Japan Atomic Energy Research Institute. As a result, the Westinghouse computer codes were validated as sufficient for use in performing accident analyses in accordance with the requirements of 10 CFR Part 50 and Part 52. In addition, the NRC performed independent analyses of the AP600 using different analysis codes to confirm the adequacy of the AP600 design as well as the AP600 safety analysis presented in the AP600 Standard Safety Analysis Report. These independent analyses also confirmed the large safety margins exhibited in the AP600.

Westinghouse is developing a larger version of the AP600 called the AP1000. The AP1000 design is based largely on the AP600. It employs passive systems that operate in the same manner as the AP600 passive systems. The AP1000 is being designed to meet NRC's regulatory criteria in a similar manner to that found to be acceptable for the AP600. The AP1000 is being designed to meet NRC's deterministic safety criteria and probabilistic risk criteria with large margins.

Westinghouse intends to certify the AP1000 standard plant design under the provisions of 10 CFR Part 52. To that end, Westinghouse submitted an application for Design Certification of AP1000 to the NRC on March 28, 2002. This NERI program provided support for development and analysis of some areas of the design that are included in the Design Certification application. AP1000 design features, as they relate to Design Certification, are included in the AP1000 Design Control Document (APP-GW-GL-700).

### Research Progress

AP1000 uses a canned motor pump for its reactor coolant pump. Canned motor pumps are used in the U.S. Navy's nuclear program and are part of the AP600 design. The pump and motor size required for AP1000 is an extension from current practice. The plant designers worked with the pump designers to develop a pump specification that met plant requirements while minimizing the pump design extension.

AP600 is designed and certified based upon a given version of the ASME Code and other applicable national consensus standards. AP1000 should be based upon more current versions of those standards. This NERI program partially supported a study to determine which version of the ASME Code will be the basis for AP1000 and the technical basis that was provided to NRC to justify use of this version.

The steam generators for AP1000 are larger than those for AP600 and are based upon the replacement units for Arkansas Nuclear Unit 1. A unique specification was prepared to account for the AP1000 thermal hydraulic operating conditions and for the channel head mounted reactor coolant pumps.

Safeguards data packages were prepared to support safety analyses and the safety analyses themselves were performed. This NERI program helped support analysis in two areas. The results of all safety analyses are included in the AP1000 Design Control Document. The Phase 2 report for this NERI program provides examples of the nature and results of the two supported efforts for AP1000.

In 2002, the NRC prepared and transmitted 700 Requests for Additional Information (RAIs) to Westinghouse. This completes the generation of RAIs and Westinghouse will have answered them all by December 2, 2002. A large number of these RAIs relate to safety analysis. They request additional information on code development, analysis techniques, model development, assumptions, relevant equations, applicability of tests, and other details. There are no RAIs that question the basic safety of AP1000. All RAIs do require a thoughtful, documented answer for NRC to use in its development of the AP1000 Safety Evaluation Report.

In conclusion, this NERI program provided support for representative design certification activities. These activities are unique to AP1000, but are representative of research activities that must be driven to conclusion to realize successful licensing of the next generation of nuclear power plants in the United States.

## Planned Activities

In Phase 3, Westinghouse will perform the safety analyses necessary to answer Requests for Additional Information (RAIs) from the NRC. Phase 3 activities will be in the areas of Loss of Coolant Accident (LOCA) analysis, non-LOCA analysis, and containment response analysis. A final report will be issued outlining the NERI supported safety-related analysis performed for AP1000.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Design and Layout Concepts for Compact, Factory-Produced, Transportable, Generation IV Reactor Systems

Primary Investigator: Fred Mynatt, University of Tennessee

Collaborators: Massachusetts Institute of Technology; Westinghouse Electric Company LLC; Oak Ridge National Laboratory; Newport News Shipbuilding; Institute for Physics and Power Engineering

Project Number: 00-047

Project Start Date: August 2000

Project End Date: September 2003

## Research Objectives

The purpose of this research project is to develop compact Generation IV nuclear power plant design and layout concepts that maximize the benefits of factory-based fabrication and optimal packaging, transportation, and siting. The potentially small footprint of Generation IV systems offers the opportunity for maximum factory fabrication and optimal packaging for transportation and siting. Barge mounting is an option to be considered and will offer flexibility for siting including floating installation, on-shore fixed siting, and transportation to nearby inland sites. Railroad and truck transportation of system modules will also be considered in this work. The project utilizes the work of others, including both previous efforts and current Generation IV work. This includes a previously funded NERI project to develop standards and guidelines for cost-effective layout and modularization of nuclear power plants.

## Research Progress

This project was funded as a grant to the University of Tennessee (UT) and subgrant to the Massachusetts Institute of Technology (MIT) with a starting date of August 15, 2000. Following the grant processing and assignment of students, work began on the first tasks—acquisition and reviews of available designs and requirements for each reactor type. Based on that work, the project team selected three reactor types. These included a helium-cooled Modular Pebble Bed Reactor (MPBR) concept being developed at MIT, the International Reactor, Innovative & Secure (IRIS) water-cooled concept being developed by a team led by Westinghouse Electric Company, and a lead-bismuth-cooled concept to be developed by UT. Work began about February 15, 2001, on computer modeling of the reactor systems and initial plant layout concepts were completed by October 15, 2001.

Development of plant layout and modularization concepts requires an understanding of both primary and secondary systems. Work to develop the MPBR at MIT included the initial concepts for both systems. The IRIS project did not have a secondary system conceptual design nor were appropriate primary and secondary system concepts available for a lead-bismuth cooled reactor. During the second phase of the project, efforts were focused to further develop the MPBR concept, and to develop a secondary system and integrated plant concept for IRIS and a lead-bismuth-cooled integrated plant concept. There was also increased interaction in the second phase among researchers on the IRIS development team for the light water reactor (LWR) concept, those working on the Oak Ridge National Laboratory MPBR concept, and several other individuals also working on lead-bismuth-cooled reactors.

LWR Concept: The objective of this work is to develop a conceptual design and layout of the balance-of-plant (BOP) for a Generation IV nuclear power plant using the 300-MWe Westinghouse IRIS as the primary system. The motivation to develop the BOP concept is to create a layout for use in modularity and manufacturing studies. In order to determine the layout, various requirements must be taken into account, for example, standard design specifications. The research focuses on two aspects: conceptual design of the BOP Power Conversion systems for the IRIS reactor, and layout of the design concept in a 3-D Computer Aided Design (CAD) package. This will provide the information needed to carry out modularity and manufacturing simulations and design specifications of the nuclear power plant.

The preliminary, conceptual design and balance of plant for a Generation IV nuclear power plant using the Westinghouse IRIS has been developed. This plant design formed the basis of a Master of Science thesis in Nuclear Engineering at the University of Tennessee (UT) in May 2002. The system output is 1000 MW thermal and approximately 360 MW electric. It has six feed water heaters and dual reheat with one high pressure and one low-pressure tandem compound turbo-generator unit. Sizes and weights of the components and associated piping are estimated. The final layout of the plant has a footprint that is 100 meters long by 40 meters wide, and weighs approximately 7400 tons. Figures containing visualizations of plant components layout and solid modeling have been developed. The main constraints for modularity are to section the plant for barge transport,

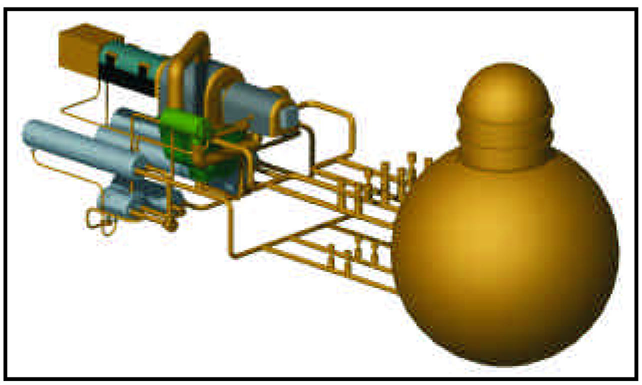


Figure 1. The figure is a visualization of IRIS containment and BOP components.

which requires that the sections must fit on a barge no larger than 400 feet long by 110 feet wide with a draft less than 9 feet. These limitations are dictated by water depth (9 feet minimum channel depth at mean low water) and specified lock sizes from the mouth of the Mississippi River northward to the Ohio River, for possible siting at Portsmouth, Ohio. The present preliminary plant design meets these criteria. Optimization of plant components to maximize efficiency and permit maintenance and repairs while minimizing capital costs has not been accomplished, although efforts to do so have been initiated for the feed water reheat system. Figure 1 shows a three-dimensional model of the IRIS containment and BOP components.

<u>MPBR Concept:</u> The MPBR concept project interfaces closely with another NERI project at MIT that involves designing the major components, including the intermediate heat exchangers, turbines, compressors, recuperators, precoolers, and intercoolers. The two teams are working together to establish layout options given the actual conceptual designs being developed. The MPBR design power level is approximately 100 MWe.

The modularity and packaging studies performed for the MPBR BOP can be broken down into several tasks:

1. System layout and design (physical layout and packaging of the plant components).
2. System concept design for increased modularity and decreased cost.
3. Advanced component design concepts for future implementation.

The first task involves defining the physical layout of the power plant itself and any transportation issues involved in its construction. The second task is concerned with making high-level trade studies of the actual system, such as the number of intercoolers, limiting temperatures, and other system parameters. The third task involves searching for advanced component concepts that may help with the other two tasks by making individual components simpler, cheaper, or more fault-tolerant.

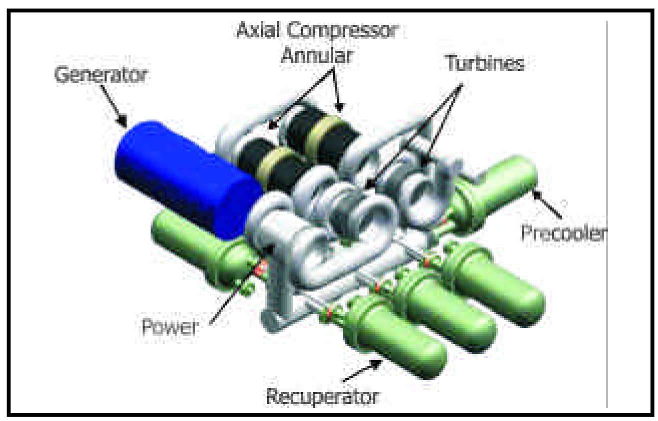



Figure 2: The diagram is a three-dimensional model of the MPBR balance-of-plant.

The proposed modular BOP system uses components and component carriers sized to fit within the limitations of truck transport. These component carriers are steel space-frames that encapsulate each component. Using this method, all the BOP components can be built in a factory and easily assembled on-site without the need for significant cranes, assembly jigs, alignment, and other special tools. Using the integral matrix structure of steel space-frames, all the necessary access hardware (e.g., catwalks, valving, flanges) can be built in the factory, further minimizing on-site assembly.  Figure 2 shows a three-dimensional model of the MPBR BOP.

The second year of work focused on the development of a modular approach to the design and construction of a pebble bed reactor at the Massachusetts Institute of Technology. The basic design of the plant remains the same. New modularity features have been identified in the area of refueling systems and in integrating the basic concept with a construction plan. More detailed understanding of the component designs and sizes has affected the layout proposed. Further refinements will undoubtedly become necessary as stress calculations on piping systems and auxiliary systems are considered (instrumentation, monitoring, and control). At this point, the modularity concepts proposed still appear to be very practical and possible.

Included are several new areas of investigation regarding the modularity concept being developed. Consideration has been given to shipment of the reactor vessel, the modularity design of the online refueling system, spent fuel storage tanks, inventory control system and costs of the components of the plant including the intermediate heat exchangers, recuperators, turbines, generator, and precooler. In addition, a construction deployment plan has been developed with a vision for the actual building and for manufacturing the components of the power plant in a virtual factory with a "just-in- time" delivery system to site.

<u>Lead-Bismuth (PbBi) Concept:</u>  Liquid metal breeder reactors hold particular promise for future energy supply since they offer an essentially infinite-time solution to energy production through effective utilization of fertile and fissile materials.  They also can be used to recycle nearly all of the radioactive waste produced by current nuclear reactors, subsequently using the waste for energy production.  Many breeder reactors have been designed and a few have been built and operated.  However, most designs have an inherent problem with positive coolant voiding reactivity coefficients, and may present more risk than many scientists would prefer to accept.  Results from calculations performed indicate that proper choices of thorium, plutonium, and uranium fuels, along with some changes in geometry, permit a PbBi cooled reactor to operate with a negative PbBi voiding reactivity coefficient, so that a reactor with considerably more inherent safety than previous designs can be designed and operated.

One significant advantage of PbBi as a coolant is that the reactor spectrum is relatively hard, which permits significant quantities of actinides to be used as fuel and eliminates the need to dispose of them as waste.  The nuclear characteristics of this design also permit operation for at least five years without refueling, or reshuffling, since the conversion ratio can be maintained very near unity.  The time between refueling is limited by performance of fuel materials rather than by the ability to sustain the chain reaction.  Proliferation resistance is improved relative to the reactors in current commercial use since the Pu-239 inventory can be held constant or be diminished, depending on fuel management choices. Figure 3 shows a three-dimensional drawing of the PbBi concept.

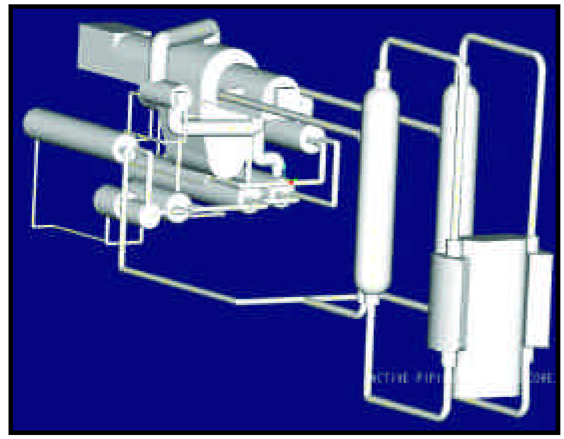


Figure 3.  The figure is a three-dimensional rendition of the lead-bismuth concept.

The reactor components in the proposed design are limited in size to a range that will allow the reactor vessel be transported on a standard rail car.  This limits the height and width to about 12 feet, the length to about 80 feet, and the weight to about 80 tons.  This should be adequate for producing 300 to 400 MW of electricity, but will depend on optimization of primary and secondary system performance, and must satisfy all licensing requirements.

Concept development and plant layout studies of a PbBi cooled reactor are completed. It was determined that a PbBi cooled fast reactor that produces 310 MWe can be designed with components that are all rail-transportable. It was further determined that a practical PbBi cooled reactor that uses only Pu as fuel, and that has a negative voiding coefficient, probably cannot be designed without the use of leakage-enhanced fuel assemblies. However, results to date indicate that a relatively high leakage slab core that uses a combination of Pu, U, and Th for fuel does have a negative coolant voiding coefficient. The reference system design uses steam generators coupled to a secondary system designed for IRIS as part of this NERI project and has an overall efficiency of about 35 percent, which could probably be increased to about 40 percent with additional design effort. A PbBi cooled fast reactor provides a long-term option for sustainable nuclear power, and it can be operated to produce very little transuranic waste.

The work on the three plant concepts and layouts has been completed. This includes refinement of the computational models and the concept layouts. The review of all three reactor concepts by Northrop Grumman Newport News (formerly Newport News Shipbuilding) has been completed. A review of the LWR concept by Westinghouse was not performed in the second phase. The DOE has approved a no-cost extension of the Westinghouse subcontract to perform related work in the third phase.

## Planned Activities

The primary effort in the third phase of this project, simulation and analysis of modular reactor fabrication and manufacturing, began on August 15, 2002. This work will be performed by an Industrial Engineering student and professor at UT with assistance and input by the professors who led the reactor concept developments. The focus is on evaluation of economies of factory fabrication versus economies of scale of site-constructed large plants. A related effort is to evaluate whether the modular approach really works in that it is feasible and cost-effective to build modules in the factory and assemble them at the site.

NUCLEAR ENERGY RESEARCH INITIATIVE

# The Secure Transportable Autonomous Reactor for Hydrogen Production

Primary Investigator: David Wade, Argonne National Laboratory

Collaborators: Texas A&M University

Project Number: 00-060

Project Start Date: October 2000

Project End Date: September 2003

## Research Objectives

The Secure Transportable Autonomous Reactor for Hydrogen production is a modular fast reactor intended for the mid 21$^{st}$ century energy market. In this system, electricity and hydrogen are employed as complementary energy carriers, and nuclear energy contributes to a sustainable energy supply based on full transuranic recycle in a passively safe, environmentally friendly and proliferation-resistant manner suitable for widespread worldwide deployment.

This is a report on progress made in the first 24 months of a three-year NERI grant to develop this concept.

## Research Progress

During the first year, the basic design selections were made and neutronics and thermohydraulics feasibility were established. The reactor is a Pb-cooled, fast spectrum, TRU-Nitride-fueled, 15-year cartridge refueled machine delivering 400 MWth of heat at 800ºC core outlet temperature. An intermediate loop (of He or $CO_2$ or molten salt) carries the heat to a Ca-Br modified UT-3[1] water cracking cycle for the manufacture of $H_2$ (and $O_2$). The water cracking cycle rejects heat at 550ºC and that heat is used in a turbogenerator to provide hotel load electricity (and optionally to provide process heat). A multi-stage flash desalinization plant receives discharge heat at 125ºC and the brine is the ultimate heat rejection from the cascaded thermodynamic cycles.

Core design work has established the feasibility of a 15-year (or even 20-year) refueling interval in a natural circulation cooled core within a rail transportable sized vessel. Passive decay heat removal has been shown to be feasible, and work on passive safety response and passive load following are targeted for deveopment and look to be favorable. Work to establish passive safety/passive load follow features will facilitate removal of all safety functions from the balance-of-plant (BOP)-a desirable feature facilitating indigenous BOP construction for job creation in developing countries.

For hotel-load electricity production, a supercritical CO2 Brayton cycle turbogenerator operating on the reject heat (at 550ºC) from water cracking was evaluated during year 2. The evaluations have shown that this cycle holds strong potential for cost reduction compared with standard Rankine steam cycle options; for example, a turbine of no more than six stages, two compressors of one to three stages each, and a finned tube economizer should achieve 45 percent conversion efficiency. The big payoff is in BOP capital cost. A 400-MWth turbine is approximately 1 m long by 2 m diameter; a reduction factor of about 50 in footprint from Rankine steam cycle equipment. Additionally, a $H_2/O_2$ combustion gas turbine was considered as an on-site peaker plant for electricity production and a bank of stationary fuel cells will be evaluated as part of the project.

A detailed evaluation was made of the S-I and the UT-3 water cracking thermochemical cycles during year 1 and a modified version of the UT-3 process has been selected. The modifications raise the theoretical efficiency to approximately 65 percent and simplify the flow sheet, offering a potential to achieve practical efficiencies in the 45 to 50 percent range in a cost-effective engineerable process. In the second year, a flow sheet and preliminary heat balance have been worked out which replace the bromine regeneration of the original UT-3 flow sheet with a plasma cracking of HBr. A detailed program of research and development has also been devised and proposed to bring the Ca-Br process to a state of readiness for a demo prototype (under separate funding). Work on reaction

[1] University of Tokyo Process

kinetics and equipment sizing to support the larger effort will be a focus for year 3.

Silicon carbide is a candidate cladding for use in high-temperature, lead-cooled nuclear reactors, and composites using silicon carbide fiber for in-vessel components are also being considered. In year 2, a corrosion test was initiated to test the compatibility of silicon carbide in molten lead at elevated temperatures (650°C to 800°C). The test uses the quartz convection loop design (shown in Figure 1) previously used in tests of various steels with lead and lead-bismuth eutectic at 550°C. A silicon carbide tube about 6 inches long and 0.5 inches in diameter was placed in the hot leg of the test loop and maintained at about 800°C while the other vertical leg was kept at about 650°C, providing for slow circulation of the lead by natural convection. The system was maintained in this condition under very low oxygen conditions for 1,000 hours (~40 days). At the conclusion of the test period the SiC showed no evidence of attack and strong evidence of nonwettability of the SiC surface by 800ºC Pb (see Figure 2).

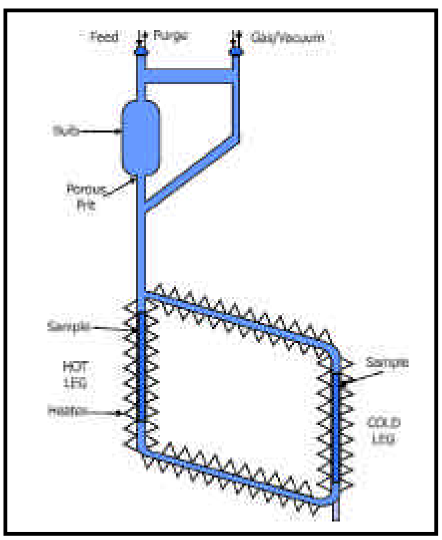
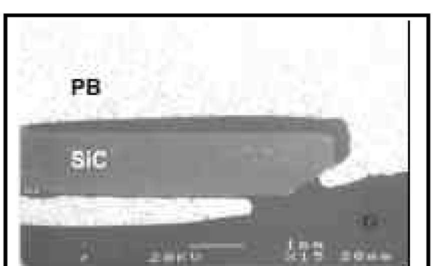



Figure 1. The schematic diagram shows the heavy metal corrosion test assembly.




No Evidence of lead interaction with SiC sample

Figure 2. Experimental results show no evidence of lead interaction with the SiC sample.

## Planned Activities

Although further optimization of core layout to flatten power density profile and increase average discharge burnup should be possible, the focus of reactor core design in year three will shift to design of thermostrucutral reactivity feedbacks to achieve passive load follow/passive safety. The heat balances of the several systems (reactor, heat transport, water cracking plant, SC-$CO_2$ cycle, and desalinization plant) will be integrated into an overall plant design; a load schedule will be developed consistent with the passive load follow/passive safety strategy, and a plant dynamics model will be created and used for safety analyses.

Contacts have been made with experts in the commercial optimization of desalinization equipment and with a U.S. utility already engaged in water desalinization. With their input, there are plans to evaluate hybrid options (flash evaporation/reverse osmosis) as alternatives to the current reference (flash evaporator) desalinization bottoming cycle.

Using corrosion test loop equipment emplaced in year 2, materials screening will be conducted in 800ºC Pb of additional structural material candidates (e.g., ZrC, ZrN, vanadium alloys, or others).

NUCLEAR ENERGY RESEARCH INITIATIVE

# Development of Design Criteria for Fluid Induced Structural Vibrations in Steam Generators and Heat Exchangers

Primary Investigator: Ivan Catton, University of California, Los Angeles

Project Number: 00-062

Project Start Date: August 2000

Project End Date: September 2003

## Research Objectives

Three objectives were defined for Phase II of this project whose goal is to develop degisn criteria for fluid induced structural vibrations in steam generators and heat exchanges:

(1) Single Phase Flow: To measure velocities and pressure distributions in square arrays with different pitch-to-diameter ratios for rigid and flexibly mounted tubes. Several heated tubes to be included to determine the relationship between instability avoidance and heat transfer penalty. Determine the entrance length and develop a stability map.

(2) Two Phase Flow: To determine the fluid-elastic instability conditions on tube arrays held at different orientations to the flow. Based on the data, develop an instability map. Conduct single tube steam-water flow and stability tests and compare results to air-water data. Measure the flow and when the tubes are heated to cause boiling. Establish the differences between air-water and steam-water flow and stability characteristics.

(3) Theoretical Development: Develop numerical algorithms for solving the governing equations using the vorticity transport equation as a first approximation. Use data from measurements of pressure differences in an array of rigid cylinders to check the validity of pressure variation assumption. Develop an energy equation for use in steam-water studies. Begin the development of models to describe the instabilities. Initiate development of tools to model the entrance region. Improve the constitutive relations and models needed for flow and stability modeling. Incorporate basic design data (tube material, diameter, wall thickness, length, type of support, and internal flow and heat transfer) into the models. Develop a relationship between tube supports used in experiments and prototypic supports.

The deliverables and milestones will include reporting experimentally based, single-phase entrance region flow and stability maps. Air-water stability maps will be developed and reported, and the impact of tube boiling will be reported. A decision about the level of detail needed in experimental studies of steam-water will be made. A report will be generated on the comparison of predictions with experimental results, and areas of weakness will be delineated; on the importance of prototype design variables in conducting laboratory tests of stability; and, on the results from evaluation of the impact of vibration avoidance on heat transfer.

## Research Progress

Progress made on each objective will be reported in turn.

Single-phase Flow: The focus of the experiment has been shifted to the measurement of real-time, two-dimensional displacement of the tubes, rather than the measurement of the flow velocity field and pressures distributions around the tubes. The reasons for this shift is to focus attention on the primary result of instability, that of tube displacement. Further, tube displacement will be compared to model predictions. Though the eventual measurement of the velocity and pressure fields are essential, the top priority is to first validate the experimental design and construction by measuring the tube-displacement magnitudes for the range of velocities of which the system is capable. When fluid-elastic instability is achieved (as determined by the reduced velocity to vibration correlation) more detailed measurements can be made.

In designing and constructing a single-phase flow loop for supplying water at room temperature, the salient features of the loop include the following (see also Figure 1):

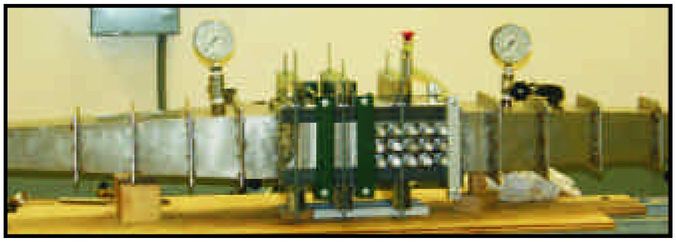


Figure 1. The photograph shows the single-phase test section and diffusers.

- A 15 hp centrifugal pump to supply 150 gpm of water at atmospheric temperatures
- A water accumulator ahead the test section in order to damp out the vibrations in the fluid
- A differential pressure flow meter

The experimental test section consists of the following:

- Inlet diffusers to avoid flow separation and flow straightener to make the flow uniform
- Acrylic box where a 5x3 dummy array of fixed tubes and a 5x3 array of flexible tubes are arranged in a normal square pattern
- Outlet diffuser
- External structure designed and installed to reinforce the test section and the diffuser against bending
- Pressure gages installed ahead and after the section

The flexible array of tubes consists of stainless steel tubes, 9 inches long and with a 1 inch outer diameter, suspended from the walls of the test section using stainless steel piano wires. A tensioning mechanism composed of high precision compression springs allows the user to change the tension in the piano wires and therefore the natural frequency of the tubes. The test section is optically accessible: a non-intrusive measurement system has been used to observe the tube motion.

The Data Acquisition System consists of the following:

- Two high-speed (120 frames per second) high-resolution (640 x 480) cameras
- Two computers ( 1.2 GHz processor, 1 Gigabyte RAM memory) dedicated to image acquisition

Preliminary results include the following:

(1) The test section has been assembled and tested.

(2) The array of flexible tubes has been calibrated, using one camera and a strobe lamp at a frequency of 16 Hz in still water (see Figure 2).

(3) Movies have been taken of the tubes at different flow rates.

(4) During the current development of methodology for data reduction, tresholding, filtering, subtraction, and cross-correlation of images techniques are being used to determine tubes displacements and orbits.

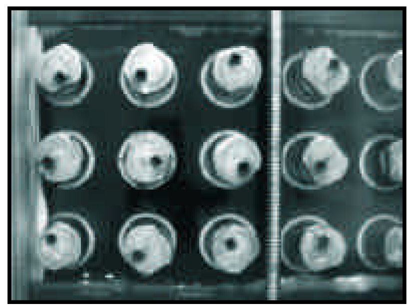


Figure 2. The photograph shows the camera output image with the array of vibrating tubes.

(5) Implementation of image processing codes has been accomplished using software such as Java Image, Adobe Photoshop and Matlab.

<u>Two-phase:</u> Design and construction of a two-phase flow loop for supplying both an air-water and steam-water mixture has been completed. The salient features of the loop include the following (see also Figure 3):

- A 10-hp centrifugal pump to supply 70 gpm of water at atmospheric pressures and temperatures
- A 46-kW electric boiler to generate steam

The experimental test section features a 5x3 dummy array of fixed tubes and a 5x3 array of flexible tubes arranged in a normal square pattern. The stainless steel

Figure 3. Pictured are the two-phase flow loop and test section.

tubes are 8 inches long and have an outer diameter of 0.625 inches, and are suspended from the walls of the test section using stainless steel piano wires. Strain gages mounted on shims welded to the piano wire are used to measure the natural frequency and damping coefficient of the tubes as they vibrate. A half bridge configuration of strain gages is used to compensate for change in length of the piano wire due to temperature.

Preliminary results of the two-phase flow include the following:

(1) The strain gages have been successfully used to calculate the natural frequency of vibration of the tubes in air and water, as well as the damping coefficient ($\zeta$).

(2) These two quantities have been calculated from the time history of the strain gage signal and also the Fourier transform of the signal.

(3) The natural frequency of the tubes in air is found to lie between 20-25 Hz.

(4) A damping coefficient of 1 percent is observed in air.

(5) Unstable motion of the tube characterized by large amplitude vibrations beyond a particular value of the air flow rate has been observed in air-water flow.

(6) Movies have been made of the instability. The strain gage signal also shows the instability as shown Figure 4.

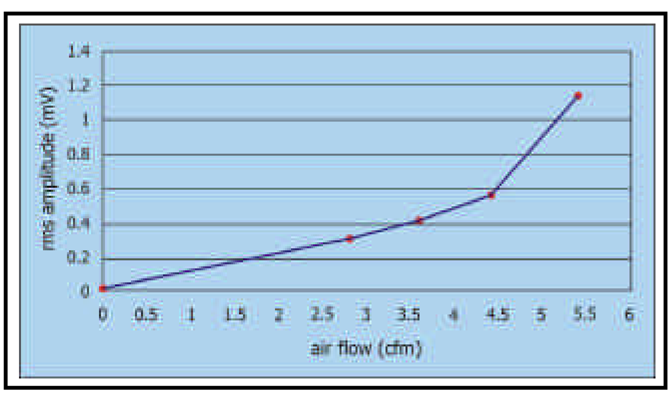



Figure 4. The graph illustrates fluid-elastic instability in air-water flow (water flow rate held constant at 32 gpm).

## Planned Activities

For the single-phase effort, more experiments will be conducted with the existing experimental setup to determine the relationship between the RMS displacements of each of the tubes in the array as a function of the flow speed, so that instability maps can be drawn. The image-processing program will be developed and used for preliminary analysis of the experimental data; using cross-correlation techniques between subsequent images, it will be possible to measure the tube displacements as well the direction of motion. Additional properties of the tube vibrations, such as relative tube motions, orbits followed by the tubes, and statistical properties, will be computed from the displacement data.

In the two-phase experiment, after successful demonstration of fluid-elastic instability using a single tube in two-phase flow, the entire 15-tube array is being installed in the test section. Once this is done, baseline tests will be made for the instability in single phase flow. The results of this test will be used to determine the parameters required for the two-phase tests. Fluid-elastic instability results in air-water and steam-water flow will be obtained. An effort will also be made to determine the damping characteristics of the tube in two-phase flow. Following this, tests using a rotated square array of tubes will be carried out.

Additionally, papers have been presented and abstracts have been or will be submitted for several conferences in 2002 and 2003.

NUCLEAR ENERGY RESEARCH INITIATIVE

# An In-Core Power Deposition and Fuel Thermal Environmental Monitor for Long-Lived Reactor Cores

**Primary Investigator:** Don W. Miller, The Ohio State University

**Collaborators:** University of Akron; Westinghouse Electric Company

**Project Number:** 00-069

**Project Start Date:** July 2000

**Project End Date:** September 2003

## Research Objectives

The primary objective of this program is to develop the Constant Temperature Power Sensor (CTPS) as an in-core instrumentation that will provide a detailed map of local nuclear power deposition and coolant thermal-hydraulic conditions during the entire life of the core. In the case of the some DOE Generation IV reactor cores, this could include normal operation, post-accident operation, and monitoring after the core is placed in permanent storage. The sensors used in this instrumentation must have a lifetime comparable to the core and be compatible with the neutronic and thermal conditions expected over the range of proposed Generation IV reactor designs. Further, the sensors must be robust and capable of operation even with extensive material degradation and, if required to achieve this objective, they must provide for in situ calibration and performance monitoring.

The CTPS concept is based on the idea of maintaining the temperature of a small mass of actual reactor fuel or fuel analogue constant by adding heat through resistive dissipation of input electrical energy. A feedback control loop is used to provide the exact amount of input electrical energy needed to keep the fuel mass at a specified constant temperature, well above the coolant bulk temperature, regardless of the nuclear energy deposited in the mass. Energy addition to the fuel mass and fuel temperature feedback to the controller are both provided by a simple resistive heating element embedded in the fuel mass. The input electrical energy required to maintain a constant temperature provides a measure of the actual nuclear energy deposition, since they are inversely related.

## Research Progress

This report provides a summary of the progress made on the tasks specified for the second year of the three-year program as described in the program proposal. The remainder of this section is a short narrative that summarizes progress, key accomplishments, and significant problems encountered, and their resolution, for each of the second-year tasks.

Second Year Program Tasks: The Ohio State University

- The task, "Modification of the Numerical Model to Represent the Planar Constant Heat Flux Power Sensor (CHFPS)," was originally unplanned, but became required when OSU investigators decided to fabricate and evaluate planar screen-printed sensors. This task was completed and results are being used in the design of the planar CHFPS. (A previous task, completed in Year 1, involved modification of the numerical model to represent the cylindrical CHFPS.)
- As noted in the last Annual Report, it was substantially more challenging than expected to design and fabricate sensor prototypes that will reliably operate in the expected environment. Initially, an attempt was made to utilize a method comparable to the first prototype. However, it soon became apparent that due to both size and required operational temperature this may not have resulted in a sensor with the required performance characteristics. Consequently, a sensor was designed with a planar configuration that requires a controlled thin layer of platinum on a sensor cone comprised of uranium oxide. To obtain a layer sufficiently robust to permit the sensor to reliably operate in the expected environment has required significant development of new materials technology.

- Good progress has been made on developing the planar configuration, and based on preliminary testing, investigators believe it will be sufficiently robust to operate in the environmental conditions of the International Reactor, Innovative, and Secure (IRIS) concept. To validate this conclusion, the numerical model was modified to represent this configuration and it has been used for design and performance modeling. Simultaneous progress was made on the original cylindrical configuration and it is now believed that both configurations will have a high probability to operate successfully in the challenging environmental conditions posed by the IRIS. Operational prototypes of both configurations are expected by the end of August.
- A critical test during the fabrication process of both sensors is evaluation of the physical integrity of components and contacts at temperatures above expected operational temperatures, therefore providing a preliminary test of sensor materials.
- As discussed in the Annual Report, the Test Digital Controller and Thermal Monitoring System was extensively tested in the first year. It will continue to be tested during completion of second-year tasks.
- In the task to develop and evaluate in-situ performance monitoring, both compensation and fault detection will be used to identify any discrepancies between the actual sensor and a sensor model. The process of compensation uses the sensor output and the sensor model to estimate the environmental temperature and the heat transfer coefficient. Fault detection will be used to obtain current sensor characteristics by measuring both the sensor input and output. A difference between the measured dynamics and the model indicates a fault. This protocol has been evaluated with the numerical model and will be used as the basis for an on-line monitor of CTPS performance, which will be evaluated experimentally during the third year of the program.
- One of the first tests of both the planar and cylindrical sensors will be a short-term radiation exposure that provides a preliminary radiation test of sensor materials. As discussed in the Annual Report for the first year, a potential problem was identified related to long-term testing, which was to begin in the second year. In preparation for this task, OSU investigators had several discussions with the staff at the Ford Research Reactor at the University of Michigan and determined that this facility will meet program needs. However, it should be noted that the Ford reactor is scheduled for permanent shutdown, pending a review by the Department of Energy for additional funding.

Second Year Program Task: Westinghouse Electric Company

- Investigators at Westinghouse have started work on the task to evaluate the effects of the sensors on the reactor environment. The selection of the first reference core configuration for IRIS is nearing completion. The sensor dimensions, bulk materials, and quantities and isotopic mixtures of fissionable materials will be used with the currently available information on IRIS core configuration to evaluate the perturbations on the neutron and temperature environment resulting from the presence of CTPS and CHFPS units. This task will be completed by the end of August 2002.
- The Year 2 milestones of completing characterized prototype sensors and controllers, and identifying effects of in-core monitors on the reactor designs will be completed by the end of the second month of the third year. Although this is two months behind the planned schedule, the Westinghouse research program is in a good position to complete the program as planned. More importantly, a sensor configuration—a planar sensor with screened printed electrodes—will be available that is likely to be superior to the planned cylindrical sensor configuration.

## Planned Activities

The following milestones have been established for the third year of the program. Selected activities to attain each milestone are described after each respective milestone. A detailed test plan is currently being developed.

(1) Complete long-term irradiation and thermal cycle testing. Cylindrical and planar sensors will be placed in the University of Michigan Ford Research Reactor in a location with maximum fluence over an approximate twelve-month time period. The physical integrity and electrical output of the sensors will be monitored.

(2) Complete numerical model validation. A static calibration will be completed of cylindrical and planar sensors over as wide a dynamic range as possible in the OSU Research Reactor. Additionally, a dynamic calibration of cylindrical and planar sensors will be completed over as wide a dynamic range as possible in the OSU Research Reactor. This will require measurement of the sensors' transfer functions using a local flux oscillator and neutron noise analysis, scram response, and response, following various positive reactivity insertions. Calibrations and testing will take place over a range of temperatures from ambient through 800°C.

(3) Evaluate in situ calibration algorithms. Performance evaluation will take place during numerical model validation and other operational scenarios.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Design and Construction of a Prototype Advanced On-Line Fuel Burn-up Monitoring System for the Modular Pebble Bed Reactor

Primary Investigator: Bingjing Su, University of Cincinnati

Collaborators: North Carolina State University; Massachusetts Institute of Technology (MIT)

Project Number: 00-100

Project Start Date: August 2000

Project End Date: September 2003

## Research Objectives

The overall goal of the project is to conceptually design, construct, and test an advanced on-line fuel burn-up monitoring system for the next-generation Modular Pebble Bed Reactor (MPBR). The MPBR is a high-temperature, gas-cooled nuclear power reactor currently under study as a possible Generation IV system. In addition to its inherently safe design, a unique feature of this reactor is its multi-pass fuel cycle in which the graphite fuel pebbles are randomly loaded and continuously cycled through the core until they reach their prescribed End-of-Life burn-up limit (~80,000 MWD/MTU). Therefore, an on-line measurement system is needed to accurately assess whether a given pebble has reached its End-of-Life burn-up limit and thereby provide an on-line, automated go/no-go decision on fuel disposition on a pebble-by-pebble basis.

This project investigates approaches to analyzing pebble bed fuel in real time using gamma spectroscopy and possibly using passive neutron counting of spontaneous fission neutrons to provide the speed, accuracy, and burn-up range required for the MPBR. The project involves all phases necessary to develop and construct a burn-up monitor, including a review of the design requirements of the system; identification of materials and methodologies that would satisfy the design requirements; modeling and development of potential designs; and finally, the construction and testing of an operational detector system.

## Research Progress

The project is to be conducted in three phases. Phase 1 is designed to characterize the fuel burn-up and radiation of fission products and has three specific tasks:

A) Identification of the system requirements
B) Fuel depletion modeling
C) Radiation/burn-up correlation analysis

The objective of Task A is to determine the design requirements for an on-line burn-up monitoring system to be used with the MPBR plants. This task involves collecting the most recent design parameters from the MPBR design effort underway at MIT, the Idaho National Engineering and Environmental Laboratory, and South Africa's ESKOM[1]. Based upon the information collected, the required availability of the burn-up monitor, pebble throughput rates, burn-up ranges of interest, cool down time for pebbles prior to measurement, and other basic physical parameters that are needed for the on-line burn-up monitor design have been determined. The objectives of Tasks B and C are to accurately model the buildup of fission products and transuranic elements in irradiated fuel pebbles and to identify correlations between types, amounts, and spectra of radiation emitted from a fuel pebble and the fuel burn-up level of the pebble. To this end, the ORIGEN2.1 code was used to perform the fuel depletion calculations and the utilization of passive gamma-ray spectrometry and neutron counting methods was investigated to establish a power-history and cooling-time insensitive burn-up measurement approach, which relies on the relationship between the burn-up and the radiation emitted by the fuel pebble from fission products and heavy actinides that have built up during its passing through the core.

In the case of gamma-ray measurements, it was found that accurate and predictable correlations between activity and burn-up can result if Cs-137 (~30.2 years half-life) and/or Eu-154 (~8.5 years half-life) are used as indicators. The correlation is linear in the case of Cs-137

[1] Electricity Supply Commission

and fourth order in the case of Eu-154. The activity of these radionuclides exhibited a behavior that is highly independent of the cooling time and the in-core power history (i.e., the pebble's path in the core), which resulted in variations in the predicted discharge burn-up of approximately 3 to 5 percent when the cooling time was varied between 0 and 7 days and the power was varied between 50 percent and 150 percent of the nominal thermal core power. However, for Cs-137 the activity measurement will be performed using the 662 keV gamma line, which is significantly interfered by the 658 keV line of Nb-97. This spectral interference cannot be completely resolved even when using germanium high-resolution detectors.

In addition, the option of using artificially introduced dopants (e.g., Co) as burn-up monitors was studied. The use of such dopants may be desirable because they can provide intense and high-energy gamma rays that have improved signal to noise characteristics, and therefore, can improve the accuracy of the measurement. Results indicate that the relative activity ratio of Cs-134 to Co-60 is somewhat more resistant to power variation and thus is a potential indicator of discharge burn-up that is accurate to within 5 percent. Furthermore, the use of a relative burn-up indicator may eliminate the need for an absolute efficiency calibration of the gamma ray detector and thus minimize the contribution of the detector calibration error in the final uncertainty analysis of the burn-up monitoring system.

The use of passive neutron counting has also been investigated as a method for burn-up measurement. In this case, the neutrons are generated either by the spontaneous fission of heavy actinides or from the ($\alpha$,n) reactions that take place within the pebble. The depletion calculations indicate that the ($\alpha$,n) component is negligible compared to the contribution of spontaneous fission. Moreover, the spontaneous fission component is dominated by the contribution from Cm-244 that has a half-life of 18.1 years. This allowed the establishment of a correlation between the total number of neutrons emitted by a pebble and its calculated burn-up, using a fourth order polynomial. Moreover, the total number of neutrons emitted is substantially insensitive to the cooling time and the in-core power history, which resulted in variations in the discharge burn-up prediction of less than 10 percent when the cooling time varies between 0 and 7 days and the power varies between 50 percent and 150 percent of the nominal thermal core power.

Although the work of Phase 1 concludes that passive gamma-ray spectrometry of selected fission products and passive total neutron counting have the potential to be developed as credible methods for on-line burn-up measurement, the feasibility of using any of the above approaches must take into account other considerations. Paramount among these is the ability to perform the measurements within the realistic requirements of throughput and the overall system reliability. That is the focus of Phase 2 efforts, with the objectives of establishing signal-to-source response functions for candidate detector systems, detector selection and optimization, and conceptual system design. To date, the following has been accomplished:

(1) The signal-to-source response functions have been simulated by using MCNP for several candidate gamma ray spectrometers that are based on either cryogenically cooled or room temperature HPGe detectors. A typical result is shown in Figure 1. In addition, the Gaussian

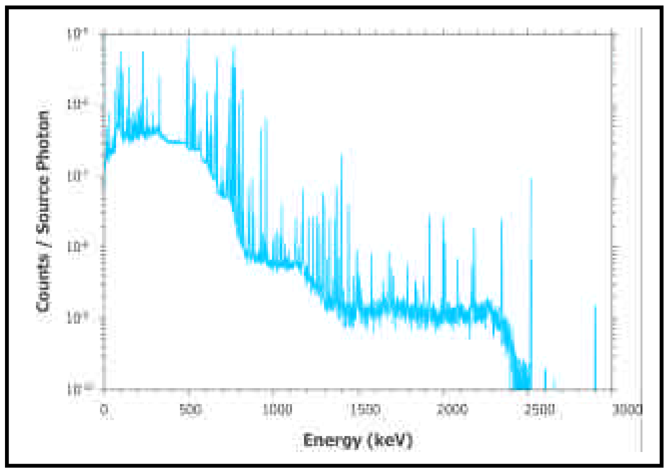



Figure 1. MCNP simulated gamma spectrum from a pebble at a burn-up of 80,000 MWD/MTU

broadened spectra for these detector responses are produced by using the FWHM vs. Energy relation generated by SYNTH.

(2) The minimum detectable activities of candidate fission products were calculated for gamma measurements. The results indicate that for the gamma spectroscopy, with approximate values for the detection efficiency, the expected measurement time that would allow a statistically accurate result for burn-up measurement varies between 30 to 60 seconds. This will be sufficient to meet a circulation rate of one pebble every 30 seconds, especially if a multi-detector system with enhanced detection efficiency is used.

(3) Detection efficiency for neutron counting has been simulated by MCNP for several common neutron detectors, including gas recoil counters, fission chambers, and passive neutron assay by the $(n, \gamma)$ conversion. Due to the low emission rate of spontaneous fission neutrons, only the BF3 long tube counter is found to be able to register a statistically meaningful neutron count within 30 to 60 seconds, if all background noises are neglected.

(4) The interference from gamma rays to a BF3 neutron counter was also simulated by MCNP. Results show that pulses (in 2 to 3 MeV) produced by gamma rays in the detector overwhelm the neutron pulses, due to the extremely high ratio ($> 10^9$) of gamma emission rate over neutron emission rate. Such interference cannot be overcome by using gamma shielding. Although other neutron detection schemes will be investigated in the next phase, it seems unlikely to have an accurate neutron counting within the required time interval, considering the interference from gammas. Therefore, it is unlikely that neutron counting is usable for on-line burn-up determination.

## Planned Activities

The research efforts in Phase 2 involved developing the signal-to-source response functions for candidate detector systems and identifying optional detection methods for use in the burn-up monitor. The objective in the remaining phase of this project is to finalize an optional (hopefully optimal) design for the on-line, burn-up monitor system and to test the functionality and performance of the system by laboratory experiments. To this end, the following research activities will be carried out:

- Investigating other neutron detection methods to make a conclusion about the feasibility of using neutron counting for on-line burn-up determination
- Assessing the summing effect on gamma ray spectra in gamma measurement
- Studying further the feasibility of Co-doping for burn-up measurement
- Constructing a high resolution gamma spectrometry detector system as a burn-up monitor
- Conducting several experiments at the North Carolina State University's PULSTAR Reactor Laboratory to evaluate fundamental gamma spectrometry issues that may affect the burn-up measurement

NUCLEAR ENERGY RESEARCH INITIATIVE

# Balance of Plant System Analysis and Component Design of Turbo-Machinery for High-Temperature Gas Reactor Systems

Primary Investigator: Ronald G. Ballinger, Massachusetts Institute of Technology

Collaborators: Northern Engineering & Research

Project Number: 00-105

Project Start Date: August 1999

Project End Date: September 2003

## Research Objectives

The purpose of this project is to develop systems analysis tools for the evaluation of turbo-machinery and balance of plant (BOP) power conversion in high-temperature gas cooled reactor (HTGR) systems. These tools will then be used to develop optimized power conversion systems for HTGR systems. Current concepts for HTGR systems call for modular designs with electrical output in the 110-MWe range. Key questions which must be addressed in order for such systems to be adequately evaluated include:

(1) Can a helium power turbine be developed in the 110-MWe range?

(2) Can advanced compact heat exchanger technology be used in the design of intermediate heat exchangers (for indirect cycle plants) and/or recuperators (direct and indirect cycle plants)?

(3) Can structural and materials issues be adequately characterized to allow for detailed life-cycle analysis?

(4) How do specific component designs impact overall cost?

## Research Progress

Key progress to date includes the following:

(1) A "reference" plant design was established, which can be built with existing technology. However, the thermal efficiency of the plant will be sub-optimal due to restrictions on the reactor outlet temperature.

(2) Initial designs for the turbo machinery and heat exchangers were established, including the intermediate heat exchanger. Two concepts have been identified. However, each design will require that active measures be taken to assure safe operation.

(3) Potential vendors for all of the major components were identified and initial cost estimates for these components were obtained.

(4) A steady state model was developed for the overall plant and is being used to help optimize the system configuration.

(5) The initial transient model was developed and the process of refining the individual component models is underway.

(6) A reactor model was developed for use in both the steady state and transient models.

Each of these points is discussed in more detail below.

Project Design Goals

The overall design goals of the project were established as follows:

(1) To develop a "reference" plant design that could be built today with no significant advance in technology. This design must satisfy all appropriate codes and standards and should not require a significant R&D effort on the part of the component vendors.

(2) To answer key questions related to the design of the power conversion system. These questions encompass three general categories of (1) intermediate heat exchanger design, (2) turbine and compressor design, and (3) system control.

(3) To then develop an "advanced" design which allows for prudent and achievable extensions of

technology. In this context, "prudent and achievable" means that some development effort may be needed but that this development effort should not be a significant fraction (< 20 percent) of the cost of the system.

The design team also had to make an assumption related to the nuclear island. While it was not the task of the project to "design" the nuclear island, it was necessary to choose a reference system as the starting point. For this project, researchers elected to choose the current ESKOM PBMR reactor as the reference. They believe that this reactor is a reasonable choice because the design fits with a Brayton cycle plant that is currently being designed for commercial use and therefore will be constrained by the same codes and standards as the BOP design.

The foregoing design goals imply certain constraints on the design process. The general design constraints for the design have been established as follows:

- Compliance with the ASME Boiler and Pressure Vessel Code, Section III, Class I for the primary pressure boundary which includes the reactor vessel and piping and the primary side of the intermediate heat exchanger
- Compliance with the ASME Boiler and Pressure Vessel Code, Section VIII where applicable
- Purchasable components
- Use of the ESKOM PBMR as the primary heat source

ASME Code Compliance

ASME code compliance places limits on temperature, allowable stress, and materials selection. Section III qualified materials are the most severely limited in the entire ASME code. The requirement of Section III results in an intermediate heat exchanger inlet temperature of 400°C. In addition to this temperature limit, the maximum allowable stresses at these temperatures place severe design constraints on the intermediate heat exchanger and will require that an "operating" curve for this component be established in which time and temperature for a given differential pressure (primary to secondary) accumulation are measured and accumulated to predict component life and assure safe operation. As a practical matter this means that for certain accident scenarios, one or both of the primary and secondary sides will need to be pressure-regulated.

Purchasable Components

This design constraint results in constraints due to technology limits. For the initial phase of the project this meant that there was a limit for the maximum shaft power to approximately 50Mw (70,000 shaft HP). This limit translates into limitations on the minimum number of shafts in the BOP system. This in turn will have a direct, and negative, effect on the complexity of the control system for the BOP. For the initial design this translated into a 4 shaft BOP with three compressor/turbine sets and a two-set power turbine design.

A second limit concerned the maximum allowable helium velocity. The establishment of this limit was more subjective and the team relied on discussions with various turbine and heat exchanger vendors. The graphite structure in the PBMR core results in a velocity limit of 50 m/sec due to erosion limitations. However, with the use of an indirect cycle, this limitation does not exist. Additionally, there are no sonic limitations with helium. However, based on discussions with component manufacturers, an upper velocity limit of 120 m/sec was established. This velocity limit, for a fixed system mass flow rate and temperature, places constraints on piping diameters.

ESKOM Reactor Limits

The use of the ESKOM PBMR reactor as the primary heat source carried with it all of the materials limitations for this plant. These include the consequences of using a conventional A 508 steel for the pressure vessel and piping. The design also resulted in geometric constraints related to piping configuration and layout.

The use of A 508 steel for the pressure vessel places limits on temperature of 280°C for steady state operation and 350°C for transient, short time, operation. Due to the high gas outlet temperature (850°C in the initial design), this requires that an active cooling system be in place during operation. For the chosen design, this also means that the intermediate heat exchanger (IHX) must have a cooled Section III boundary. Additionally, since the source of cool gas for the ESKOM design is the compressor outlet, the use of an IHX complicates the design. The design "fix" for this constraint is to include, as either a separate heat exchanger or as a part of the IHX, the capability for using a compressor outlet gas stream to cool a separate cooling circuit for the pressure vessel and associated piping. As a practical matter this means that, due to the piping cooling requirement, the hot sections of the BOP (to the turbine inlets) must also have cooled pressure boundaries.

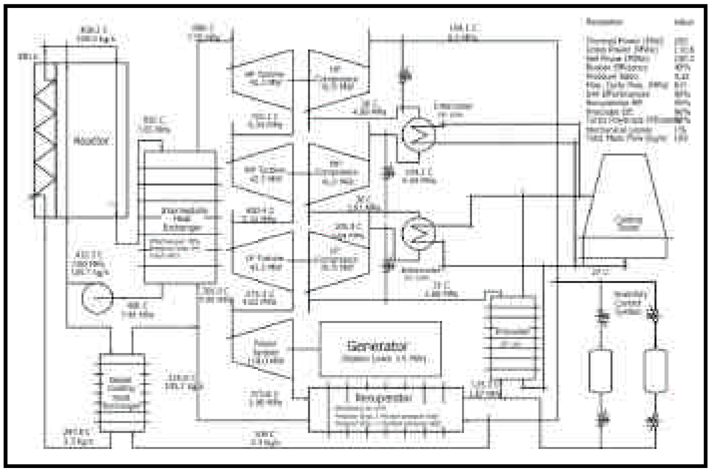



Figure 1. The schematic illustrates the reference BOP System.

<u>Current Reference Plant Design</u>

Given the initial design requirements and constraints, a reference plant has been designed and configured. Figure 1 shows a schematic of this design. Temperatures, pressures, and flow rates have been identified.

Key features of the reference system include the following:

- Use of a secondary heat exchanger for the reactor vessel and piping cooling system
- Use of both inventory and bypass control (to be discussed further below) for system control
- Location of bypass control valves on the cold side of the plant
- Use of intercooling

The reference plant is a 250 MWth design with a gross power of 110.6 MWe, which yields a net efficiency of 40 percent. The net plant efficiency is considerably less than the overall project goal of 45 percent. However, it must be cautioned that the initial phase of the program was to specify a plant with only the potential to be built using existing technology. An advanced reference design will be developed during the second year of the project that will seek to increase the overall efficiency.

## Planned Activities

The first year of the project has seen the development of the reference design, and researchers are satisfied that the design could be built, although its estimated efficiency is below the target efficiency for an economic design. An increase in reactor outlet temperature of from 850°C to 900°C will be needed to achieve this efficiency. Designs for major individual components have also been taken to the point where the team is satisfied that they could be built. In the cases of the turbines, compressors, and the IHX, it was possible to obtain initial cost estimates from commercial sources and it is believed these components could be purchased with a minimal requirement for extensive R&D effort.

The next steps in the project will be to more clearly establish the limits to the achievement of an increase in temperature. Based on this analysis, a final system design will be developed.

Although not mentioned previously in this report, work has begun on the development of a transient model during this reporting period. During the next year, the transient model will be fully developed and then used in optimization of the final design.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Forewarning of Failure in Critical Equipment at Next-Generation Nuclear Power Plants

Primary Investigator: Lee M. Hively, Oak Ridge National Laboratory

Collaborators: Duke Engineering and Services Inc.; Pennsylvania State University

Project Number: 00-109

Project Start Date: August 2000

Project End Date: September 2003

## Research Objectives

Researchers at Oak Ridge National Laboratory (ORNL) are applying new nonlinear methods to assess condition change and forewarn of machine failures from experimental test sequences. These new measures are much more discriminating than those previously applied. The specific application is critical equipment in next-generation nuclear power plants. Test data have been provided by two collaborating institutions, namely Duke Engineering and Services during fiscal year (FY) 2001, and the Pennsylvania State University during FY 2002. If successful, this effort will overcome one of the major present hurdles to automatic, timely, and reliable prognostication for condition-based maintenance and repair.

The methodology is multi-tiered, model-independent, and data-driven. The first tier rejects data of inadequate quality. The second tier removes signal artifacts that would confound the analysis, but leaves the relevant nonlinear dynamics essentially unaffected. The third tier converts the artifact-filtered, time-serial data into a phase-space (geometric) representation, which then is transformed to a discrete distribution function (DF). The nominal-state DF is compared to subsequent test-state DFs via dissimilarity measures of condition change, which are much more sensitive than conventional statistics or traditional nonlinear measures. Thus, the discriminating power of the method is less affected by noisy, finite-length datasets.

ORNL's approach yields robust nonlinear signatures of degradation, allowing earlier and more accurate detection of the deterioration onset, as well as more accurate predictions of the progress of the deterioration. Anticipation of failures in critical equipment at next-generation nuclear power plants will help in the scheduling of maintenance to minimize safety concerns, unscheduled non-productive downtime, and collateral damage due to unexpected failures. It is expected that this approach will lead to significant economic benefits and improved public acceptance of nuclear power.

## Research Progress

Long-term failure monitoring of operational equipment is not feasible within the scope of our present project, since such failures may take years to occur. Instead, data was acquired in FY 2001 from a motor-driven pump for two test sequences of progressively larger seeded faults (imbalance and misalignment). ORNL's nonlinear measures of condition change correlated well with the experimental level of vibration, both below and above the ISO 2372 and ISO 3945 limits. This work included a robust implementation of the nonlinear analysis on a desktop computer, not unlike that for acquisition and analysis at an advanced nuclear reactor. The Annual

| Data Provider | Equipment and Type of Failure | Diagnostic Data |
|---|---|---|
| 1) EPRI (S) | 800-HP electric motor: airgap offset | motor power |
| 2) EPRI (S) | 800-HP electric motor: broken rotor | motor power |
| 3) EPRI (S) | 500-HP electric motor: turn-to-turn short | motor power |
| 4) Otero/Spain (S) | ¼-HP electric motor: imbalance | 1D acceleration |
| 5) PSU/ARL (A) | 30-HP motor: overloaded gearbox | load torque |
| 6) PSU/ARL (A) | 30-HP motor: overloaded gearbox | accelerometer power |
| 7) PSU/ARL (A) | 30-HP motor: overloaded gearbox | accelerometer power |
| 8) PSU/ARL (S) | crack in rotating blade | motor power |

Table 1: Summary of PY2 Test Sequences. Table 1 also shows the type of diagnostic data that was analyzed for failure forewarning. Motor power, P, was obtained from the three-phase motor currents, Ii, and voltages, Vi, by using the formula, P =  i IiVi. The sum over i includes all three electrical phases. Accelerometer power came from tri-axial acceleration, which is a three-dimensional vector, A, that can be integrated once in time to give velocity vector, V =  A dt. Mass, m, times acceleration (vector) is force (vector), F = mA. The vector dot-product of force (vector) and velocity (vector) is power (scalar), P = F " V. The resultant three-dimensional accelerometer power captures the dynamics from all three components of acceleration.

Report, ORNL/TM-2001/195, provides details of the first year's progress, including validation of the approach on well-characterized model data.

Additional test data was obtained during the second project year, as summarized in Table 1. Some of the test sequences involved seeded faults (denoted by 'S' in Table 1), with the equipment initially in nominal operation, and subsequently with successively larger (controlled) faults, finally yielding equipment failure. A second class of accelerated failure tests (denoted by 'A' in Table 1) likewise began with nominal operation. The over-stressed equipment experienced a gradual (uncontrolled) degradation, and ultimately failed. For example, the gearbox failed by the breakage of one or more gear teeth.

ORNL's patented nonlinear measures showed clear change in every sequence, as the test progressed from nominal operation, through degradation to failure. Forewarning was not consistently seen in the conventional statistical measures, nor do traditional nonlinear measures give reliable forewarning. Figure 1 shows an example of the change in phase-space dissimilarity measures for Test-Sequence #2. This sequence began with the motor running in its nominal state. Then, one rotor bar cross section was cut 50 percent at the 11 o'clock position. Next, the same rotor bar cut completely through. Subsequently, a second rotor bar was cut 100 percent at the 5 o'clock position, and in addition to, the first rotor failure. Finally, two additional rotor bars were cut adjacent to the original 11 o'clock bar, with one bar cut on each side of the original, yielding four bars completely open. Figure 1 shows a linear rise in the logarithm of the phase-space dissimilarity measures of condition change, corresponding to the exponential rise in the magnitude of these seeded faults (successively doubling from 1/2 to 1 to 2 to 4). Research on the accelerated failure test sequences has yielded a statistical criterion that distinguishes between the gradual rise in phase-space dissimilarity measures and the abrupt (additional) increase that gives forewarning of failure. These results provide compelling evidence for failure prognostication via the ORNL nonlinear paradigm. Complete details are provided in the FY 2002 Annual Report, ORNL/TM-2002/183.

Other work during FY 2002 included actions to protect the intellectual property for this technology. Researchers responded to the U.S. Patent Office action on an earlier patent application that resulted in the allowance of all of the claims for the connected phase-space dissimilarity method. A new patent application was also submitted to protect additional improvements to the methodology that was developed more recently.

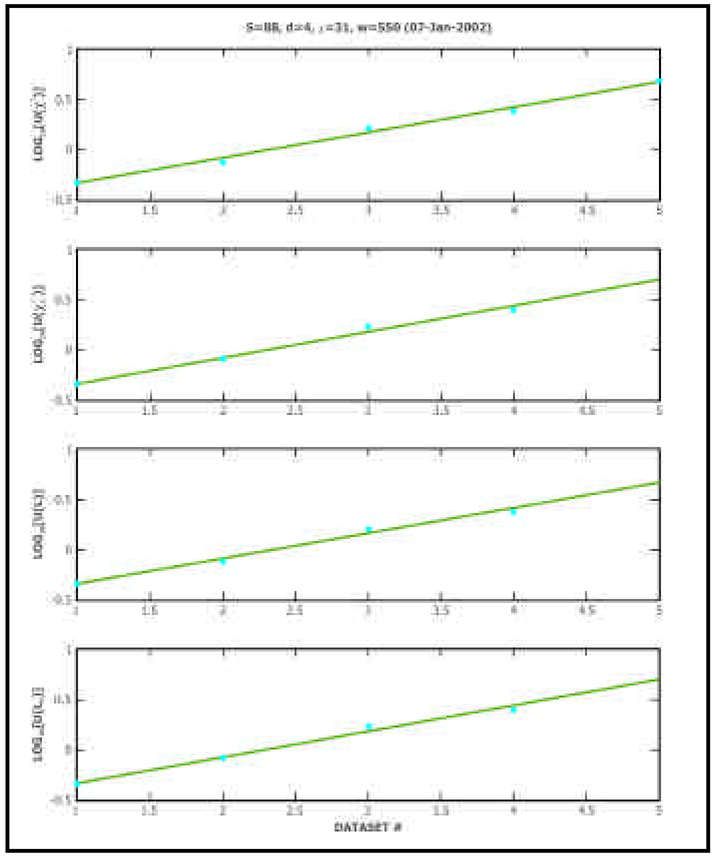



Figure 1. Plots of the four nonlinear dissimilarity measures for the broken-rotor seeded-fault power data. Dataset #1 is for the nominal (no fault) state. Dataset #2 is for the 50% cut in one rotor bar. Dataset #3 is for the 100% cut in one rotor bar. Dataset #4 is for two cut rotor bars. Dataset #5 is for four cut rotor bars. The exponential rise in the severity of the seeded faults is shown as an almost linear rise (solid line) in the logarithm of all four dissimilarity measures (*) for the chosen set of phase-space parameters.

## Planned Activities

Phase 2 work during the third project year (FY 2003) will involve acquiring and analyzing additional data from test sequences for seeded faults and accelerated failures. These tests are expected to include additional gearbox failures, cracked-shaft failures, and various generator failures in the rotor, stator, and diode. A companion effort will seek better discrimination and robustness of the forewarning methodology, along with a reduction in the analyst-intensive effort. Phase 3 work during FY 2003 will assess the usefulness of this forewarning approach in terms of failure reductions and of cost-effectiveness.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Feasibility Study of Supercritical Light Water Cooled Fast Reactors for Actinide Burning and Electric Power Production

Primary Investigator: Philip E. MacDonald, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Massachusetts Institute of Technology (MIT); University of Michigan; Westinghouse Electric Corporation

Project Number: 01-001

Project Start Date: August 2001

Project End Date: September 2004

## Research Objectives

The use of light water at supercritical pressures as the coolant in a nuclear reactor offers the potential for considerable plant simplification and consequent reduction in capital and operating and maintenance (O&M) costs compared with current light water reactor (LWR) designs. Also, given the thermodynamic conditions of the coolant at the core outlet (i.e., temperature and pressure beyond the water critical point), very high thermal efficiencies are possible for the power conversion cycle (i.e., up to about 45 percent). Because no change of phase occurs in the core, the need for steam separators and dryers as well as for BWR-type recirculation pumps is eliminated, which, for a given reactor power, results in a substantially shorter reactor vessel and smaller containment building than the current BWRs. Furthermore, in a direct cycle, the steam generators are not needed. If no additional moderator is added to the fuel rod lattice, it is possible to attain fast neutron energy spectrum conditions in a supercritical water-cooled reactor (SCWR). This type of core can make use of either fertile or fertile-free fuel and retain a hard spectrum to effectively burn plutonium and minor actinides from LWR spent fuel while efficiently generating electricity. One can also add moderation and design a thermal spectrum SCWR that can also burn actinides.

The project is organized into three tasks:

Task 1 - Fuel-cycle Neutronic Analysis and Reactor Core Design (INEEL): For the fast-spectrum SCWR, metallic (dispersion type), and oxide, fertile fuels will be investigated to evaluate the void and Doppler reactivity coefficients, actinide burn rate, and reactivity swing throughout the irradiation cycle. For the thermal-spectrum SCWR, a variety of fuel and moderator types will be assessed.

Task 2 - Fuel Cladding and Structural Material Corrosion and Stress Corrosion Cracking (University of Michigan and MIT): MIT will use an existing supercritical-water loop to conduct corrosion experiments in flowing supercritical water. To collect stress-corrosion cracking data, a high-temperature autoclave containing a mechanical test device will be built at the University of Michigan in Year 1 and operated in Years 2 and 3. The data from both universities will be used to identify promising structural and fuel cladding materials and develop appropriate corrosion and stress corrosion cracking correlations.

Task 3 - Plant Engineering and Reactor Safety Analysis (Westinghouse and INEEL): The optimal configuration of the power conversion cycle will be identified. Particular emphasis will be given to the applicability of current supercritical fossil-fired plant technology and experience to a direct-cycle nuclear system. A steady-state, sub-channel analysis of the reactor core will be undertaken with the goal of establishing power limits and safety margins under normal operating conditions. In addition, the reactor's susceptibility to coupled neutronic/thermal-hydraulic oscillations will be evaluated. The response of the plant to accident situations and anticipated transients without a scram will also be assessed.

## Research Progress

Task 1 - Neutronic Analysis and Reactor Core Design: A qualitative analysis was performed to determine which fuel form would support the highest reactivity-limited burnup in a fast-spectrum SCWR, and would have the most proliferation-resistant isotopics at a particular burn-up. A relatively long core life and a modest reactivity swing are possible in fast-spectrum SCWRs with most fuels. However, the uranium-based fuel types had the highest beginning-of-life reactivity, and the best reactivity-limited burn-up, whereas the thorium-based fuels had the best spent-fuel isotopics. Therefore, the most appropriate fuel for fast-spectrum SCWRs appears to be a mixture of thorium and uranium to balance long core life with proliferation-resistant isotopics.

In addition, the neutronic performance of several solid moderators for use in a thermal spectrum SCWR core was evaluated and compared to that of water rods. It was found that the only acceptable solid moderator is delta-phase zirconium hydride ($ZrH_{1.6}$), which generates a relatively high multiplication factor and a negative coolant void reactivity coefficient. Several issues key to the chemical and thermo-mechanical feasibility of $ZrH_{1.6}$ were assessed including zirconium-hydride/water interaction, hydrogen release, hydrogen redistribution, pressurization of the moderator box at high temperature, phase stability, and compatibility of zirconium hydride with the moderator box material. Zirconium hydride moderator rods appear to be suitable for use in SCWR thermal-spectrum cores, and therefore this approach will be pursued further during the project. Also, a simple analysis indicated that the use of zirconium-hydride moderator will not result in significant additional costs.

<u>Task 2 - Fuel Cladding and Structural Material Corrosion and Stress Corrosion Cracking Studies:</u> The design and fabrication of the University of Michigan supercritical water loop system for stress corrosion cracking tests was completed and experiments have begun (Figure 1 shows a view of the overall system). In this loop system, one tensile sample can be tested in various loading modes such as constant extension rate tension (CERT), constant load, ramp and hold, low cycle fatigue, and so forth. Additionally, six U-bend samples can be loaded into the test vessel, using sample holders secured to the vessel internal support plate. The initial test results indicate that type 304L stainless steel, which is commonly used in LWRs, is highly susceptible to stress corrosion cracking in SCWRs.

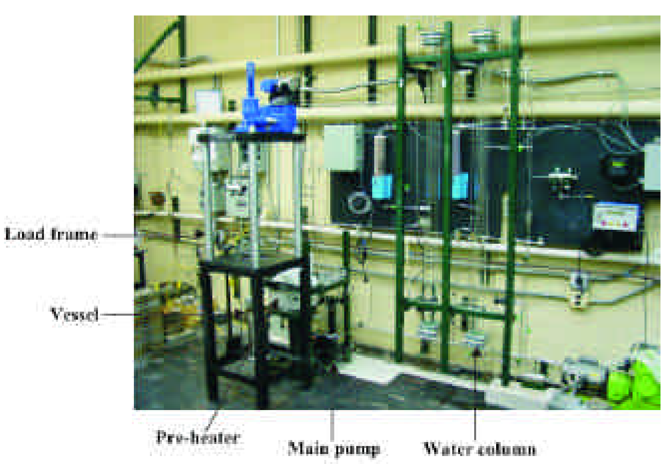



Figure 1. The photograph provides an overall view of the University of Michigan's supercritical water loop system.

The exposure facility at MIT incorporates a relatively large autoclave with an internal volume of approximately 860 ml. It is large enough to expose a rack of weight loss, welded, and U-bend samples for extended times. Initial experiments over a temperature range encompassing both subcritical and supercritical conditions have been completed with 316L stainless steel and Inconel-625 samples. Preliminary data from the Inconel-625 suggests the potential for localized breakdown and surface pitting both for exposed and occluded regions.

<u>Task 3 - Plant Engineering and Reactor Safety Analysis:</u> The preliminary core layout (dimensions, core configuration) and thermal-hydraulic design (temperature, pressure, flow rates) were developed. The design criteria for the system has been defined, and the correlations for heat transfer in supercritical water and the methods for the hot channel factors that will be used to verify the proposed design criteria for the fuel system have been identified. These criteria build on experience with LWRs, with one notable exception. Because critical heat-flux phenomena do not exist in SCWRs, it was decided that the key criterion for the SCWR core design would be the peak cladding temperature, analogous to the liquid-metal cooled fast reactor practice. Thus, a key step is having an accurate and reliable correlation for predicting the heat transfer coefficients in supercritical water cores. A review and assessment of the available correlations identified the Bishop and the Oka-Koshizuka correlations as appropriate and consistent. Three plant configurations (direct cycle, integral indirect cycle, and loop indirect cycle) have been selected for further investigation. An analysis was completed of the temperatures and density profiles in the average and hot channels for a variety of potential system configurations. Important parameters for this analysis were the core inlet and outlet temperatures and the option of canned assemblies versus an open lattice. A completely satisfactory design has not yet been identified. In preparation for the transient and accident analysis that will be conducted at INEEL in Years 2 and 3 of the project, the 3D finite-differences transient thermal-hydraulic/neutronic code, RELAP-ATHENA, was upgraded.

## Planned Activities

Plans are to complete the work described above under research objectives during the next two years. The work during Year 1 was completed on schedule.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Particle-Bed, Gas-Cooled Fast Reactor (PB-GCFR) Design

Primary Investigator: Temitope A. Taiwo, Argonne National Laboratory

Collaborators: Brookhaven National Laboratory; Commissariat a l'Energie Atomique (CEA), France; University of Rome, Italy

Project Number: 01-022

Project Start Date: September 2001

Project End Date: September 2003

## Research Objectives

The objective of this project is to develop a conceptual design of a particle-bed, gas-cooled fast reactor (PB-GCFR) core that meets the advanced reactor concept and enhanced proliferation-resistant goals of the U.S. Department of Energy's NERI program. The key innovation of this project is the application of a fast neutron spectrum environment to enhance both the passive safety and transmutation characteristics of the advanced particle-bed and pebble-bed reactor designs. The PB-GCFR design is expected to produce a high-efficiency system with a low unit cost. It is anticipated that the fast neutron spectrum would permit small-sized units (~150 MWe) that can be built quickly and packaged into modular units, and whose production can be readily expanded as the demand grows. Such a system could, therefore, be deployed globally. The goals of this two-year project are as follows:

(1) Design a reactor core that meets the future needs of the nuclear industry, by being passively safe with reduced need for engineered safety systems. This will entail an innovative core design incorporating new fuel form and type;

(2) Employ a proliferation-resistant fuel design and fuel cycle. This will be supported by a long-life core design that is refueled infrequently, and hence, reduces the potential for fuel diversion;

(3) Incorporate design features that permit use of the system as an efficient transmuter that could be employed for burning separated plutonium fuel or recycled LWR transuranic fuel, should the need arise; and

(4) Evaluate the fuel cycle for waste minimization and for the possibility of direct fuel disposal. The application of particle-bed fuel provides the promise of extremely high burnup and fission product protection barriers that may permit direct disposal.

## Research Progress

Physics calculations have been performed in support of a reference compact fast-spectrum core, based on the pebble-bed design. The study investigated the impacts of the fuel-pebble packing fraction, and fuel material form and temperature on the potential for obtaining a sustainable critical core for a long-life design. Different fuel matrix and reflector materials were also investigated. The results have provided indications that mixed uranium and transuranics (TRU) carbide and nitride fuel forms are attractive for meeting the goals of a long-life core and high temperature operation (see Figure 1). Potential matrix materials for these fuel forms are titanium nitride (TiN) and zirconium carbide (ZrC), although enrichment considerations for the nitride fuel might make carbide fuel the preferred choice. The application of both enriched uranium and weapons grade constituents of the fuel has been discarded in the current planning because of the potential proliferation issues that could arise from their use and because they generally result in a higher reactivity swing than TRU fuel.

The physics studies also indicated that the goal of a long-life core (with a conversion ratio greater than one) requires a fuel volume fraction that is larger than the one currently used in high temperature reactor (HTR) designs. A high core fuel volume fraction (of ~30 percent) or core size (power density less than 25 W/cc) is required to get the desired sustainable design with a high conversion ratio. Achieving a high fuel volume fraction would require a redesign of the typical coated fuel particles, since it implies the minimization of the volume occupied by low-density carbon buffer and SiC zones to accommodate more fuel. Calculations made under this assumption, using (TRU,U)N in TiN-15 matrix or (TRU,U)C in ZrC matrix, indicated that a high conversion ratio (of greater than 1.2) can be obtained with low reactivity losses over the 15 to 30 years of irradiation.

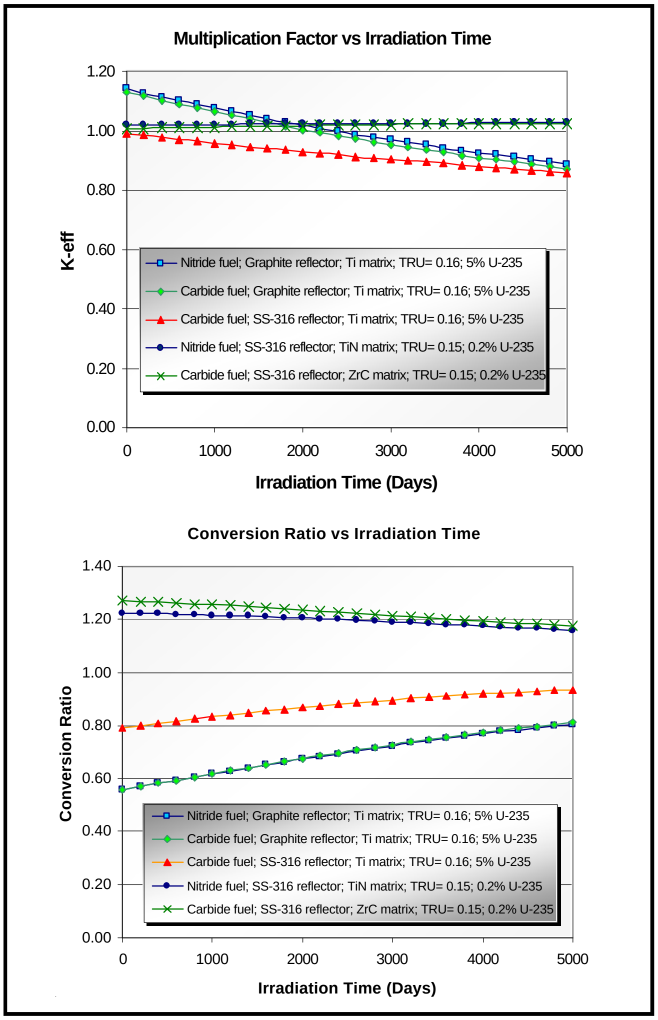



Figure 1. The graphs present the multiplication factor and conversion ratio as a function of irradiation time for PB-GCFR designs.

It is known that the safety case for fast spectrum reactors employing gas-cooling is complicated by the poor heat transfer properties and low thermal inertia of the gas coolant. The ability of this reactor type to survive a scrammed depressurization accident with a concurrent loss of electrical power, without undue hazard to the public, is clearly an attractive feature of an advanced GCFR of the Generation IV class. Researchers associated with this project have explored, and continue to examine, a number of concepts that could potentially provide this safety feature through passive means. A fundamental assessment was first performed of heat transfer modes and the implications of the decay heat curve. Scoping thermal calculations were carried out. The study revealed that natural convection at 1 atmosphere cannot be relied upon for the available selection of primary coolant gases, and that radiation through the coolant would dominate above 1,000°C. Below this temperature, a better alternative may be to provide conduction pathways. For the period immediately following scram, however, heat transfer on this timescale is not adequate for the core materials of the foreseeable future, and substantial core thermal inertia is necessary. The results also indicated that to improve the feasibility of the passive core concept, it would be prudent to reduce the reactor power envelope to below 300 MWt. Three types of basic core elements were investigated with the potential to provide core configurations that have the desired passive core safety feature: (i) block/plate, (ii) pin/tube, and (iii) pebble/particle. After the initial investigation, it was decided that the major focus of the work would be on the pebble/particle fuel element and, in particular, on the pebble-bed core configuration.

A unique concept was introduced to increase the heat storage capacity of the fuel pebble. This concept uses fuel spheres in which the center is filled with a material that does not contain fuel and which can melt and absorb heat as latent heat of fusion. This concept also substantially lowered the temperature rise within each fueled pebble. The scoping studies and the new pebble concept led to a conceptual design for an annular pebble-bed fast reactor. A severe depressurization accident was simulated for this design choice with a two-dimensional conduction model. The analysis predicted a maximum fuel temperature of 1,627°C, which is only 27°C above the current limit that is arbitrarily based on the limit for graphite fuel. The reactor vessel temperature was observed to drop sharply at the initiation of the transient, which includes a scram, and to subsequently recover to a local maximum before the final decline, which lasted until the end of the accident. The local maximum temperature of 489°C is safely below the assumed allowed value of 537°C. However, the active core power density is limited to 23 W/cc. Since this may not be economical for a fast reactor, three additional concepts were developed: (1) prompt unloading of the pebble fuel, (2) extended flow coastdown, and (3) tube reactor with a tank.

For prompt unloading of pebble fuel, during a severe depressurization accident, all of the fuel pebbles in a pebble bed reactor would be quickly dropped into a series of storage tanks. The tanks would be located inside a borated water bath. The decay heat would be conducted through the tank walls and would boil the water. The steam would rise through pipes to an air-cooled steam condenser located outside of the reactor building. A tall cooling tower over the condenser would provide a natural draft that would drive the air flow needed to cool the condenser. If the system capacity is too small to handle the entire decay heat load during the earlier part of the accident, some of the steam could be released to the atmosphere via a pressure-relief valve. The results indicate that the removal of several megawatts on a steady-state basis is quite possible with a cooling tower that is 70 feet tall and 25 feet in inside diameter at the throat.

In practice, the extended flow coastdown option is almost an instantaneous event: the reactor scram reduces the power level from full operating power down to decay levels in a fraction of a second. However, it takes many seconds for the system to depressurize and many seconds for the blowers to coast down and stop. This "coastdown" could also be extended through passive mechanisms. The ignored convective flow rate in the initial portion of the accident could potentially remove a substantial amount of decay energy and cause the predicted peak temperatures to be much lower than those predicted by the earlier analysis. With this in mind, a dynamic model was developed to include realistic pressure histories and flow coastdowns in the analysis. Results obtained showed that the results are marginal. In the case of the tube/tank reactor design, a small (long and thin) spaghetti core is proposed, composed of approximately 4 tubes (each, 0.3 meters in diameter x 4 meters in length) arranged in an array around the control rods. The tubes could be filled with fuel pebbles and would be internally cooled with high-pressure helium (~7 MPa) while the tank (calandria) surrounding the tubes would contain low pressure carbon dioxide (1 MPa) to remove decay heat at natural convection conditions. Initial calculations show that a chimney height of approximately 12 meters is required to remove approximately 1 percent decay heat.

Pursuing further the concept of combining passive conduction/radiation heat transfer with natural convection, the cold finger concept for total passive decay heat removal was conceived and developed. Cold fingers provide a passive means of decay heat removal during severe depressurization accidents. These fingers are bayonet heat exchangers, which also house the control rods and thereby serve dual purpose as both reactivity control devices and a passive decay heat removal mechanism. These fingers are inserted parallel to the axis of the core and are evenly distributed throughout its circular cross sectional area. A parametric study was performed leading to three candidate core designs that best meet steady-state thermal and hydraulic requirements for a 300 MWt gas-cooled pebble-bed reactor operating at rated conditions. Safety analyses to investigate the performance of the cold finger concept were then carried out with these core designs. The results are promising. The cold finger concept will be further explored in the second year work through the implementation of dynamic models, and dynamic analyses will be performed.

A literature review was performed to evaluate candidate fuel and materials compatibilities, high-temperature mechanical and thermal properties, and performance issues expected by operation in a fast neutron spectrum (E>0.1 MeV). Much of the effort was devoted to identifying sources of pertinent information, collecting material properties, and reviewing current gas-cooled reactor fuel designs. Space-reactor developmental efforts conducted in the 1960s were also evaluated. The literature review was completed to identify candidate fuels and materials for the development of a new GCFR to meet Generation-IV criteria.

In a similar manner, property data for industrially available structural materials with well-established manufacturing technologies were collected and assessed to identify candidate materials for various key components of the PB-GCFR. Since detailed design information about these components do not presently exist, materials and materials systems that were evaluated in other reactor-development projects were considered first. Based on this evaluation, recommendations for structural materials have been made for structures in the vicinity of the fuel zone (including ceramics such as SiC, ZrC, TiC, MgO, $Zr(Y)O_2$, TiN, and $Si_3N_4$); for the pressure vessel (2¼ Cr-1Mo and 9-12Cr steel); cooling system components (Inconel 718, Inconel 800, and Hastelloy X); shielding and thermal barriers (borated Type 304 and 316 stainless steel, ferritic HT9,and various vanadium alloys); and the reflector zone (uranium, tungsten, iron, stainless steel, and Nb-1Zr).

## Planned Activities

Physics issues that have not been investigated in detail for the long-life, PB-GCFR core will be pursued in the future. These include determining whether a single-batch fuel management scheme would require an annular core design, particularly at large core volumes, and whether enrichment splitting is required. Additionally, it is necessary to calculate core reactivity coefficients and employ them in safety analysis. Besides long-life core designs, one can envision other designs that enhance the proliferation resistance of nuclear power systems. One approach is a core design permitting the efficient and deep burning (very high burnup) of the fuel material. By using the coated-particle fuel form in the fast neutron spectrum environment, it could be possible to effectively burn the fuel. The fast spectrum is particularly suited for this task, since all TRU nuclides can be fissioned in this energy range. An evaluation of the feasibility of using the GCFR as an efficient TRU burner that enhances proliferation resistance is being planned for this project.

A detailed core design based on the cold finger concept is planned for the next phase of the project (Year 2). Heat transfer calculations for the depressurization accident with concurrent loss of electric power will be performed for this design. Additionally, when a specific core design configuration and fuel form has been selected, scoping calculations will be performed to assess the flow induced vibration and thermal stress potential of the design.

# A Miniature Scintillation-Based, In-Core, Self-Powered Flux and Temperature Probe for HTGRs

**Primary Investigator:** David E. Holcomb, Oak Ridge National Laboratory

**Collaborators:** The Ohio State University

**Project Number:** 01-039

**Project Start Date:** August 2001

**Project End Date:** September 2004

## Research Objectives

The objective of the proposed project is to develop a miniature scintillation-based, in-core, self-powered neutron flux and temperature probe. The probe would be generally applicable to any reactor technology, but would be specifically designed for the temperatures of high temperature gas reactors (HTGRs). The scintillation assembly consists of a uranium layer placed against a thick film scintillator layer. The fission fragments resulting from neutron interactions in the uranium produce light in the scintillator. The light from the scintillator is guided out from the core using a hollow core optical fiber. Both the converter layer and the scintillator are segmented. A scintillator of one wavelength is collocated with a lightly enriched $^{235}U$ uranium layer. A scintillator with a different characteristic wavelength is placed against a higher enrichment $^{235}U$ uranium layer (2 percent and 4 percent, for example). The scintillators produce different wavelengths of light, allowing independent readout of the scintillation. A conceptual layout of the probe tip is displayed as Figure 1.

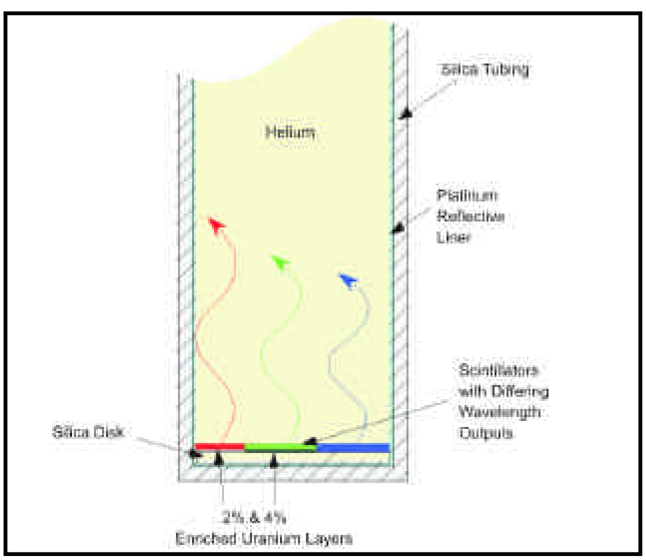



Figure 1. The graphic illustrates the conceptual layout for the scintillation probe tip.

At low temperatures (below ~400 °C), neutron flux is indicated by a weighted sum of the number of scintillation pulses produced by each scintillator (pulse mode). At higher temperatures, neutron flux is indicated by the total amount of light produced by either scintillator with the ratio of the different amount of light at the different waveband intensities serving as a burn-up monitor (current mode).

The probe indicates temperature in two manners: at higher temperatures multi-wavelength pyrometry is employed, while at lower temperatures this is supplemented by observing the optical decay time variance of the scintillation pulses with temperature. Total pulse count history is used to compensate for burn-up of the converter atoms. The detector is anticipated to have minimal gamma pulse response due to the thinness of the scintillation layer. However, the gamma response that does exist is compensated for using a scintillator layer without the neutron sensitive uranium undercoating. The thinness of the scintillator layer also minimizes the effects of the increased self-absorption (radiation darkening) with use of the scintillator material.

## Research Progress

The project began with an initial survey of the types of scintillators likely to be useful as well as phosphor layer deposition technologies. As a result of the survey, a scintillation layer deposition technique closely analogous to that employed in the manufacture of cathode ray tubes was selected and refined to the purposes of the project. Essentially, the phosphor powder is suspended in a chemically compatible liquid along with an inorganic binder. The mixture is then centrifuged down onto a substrate, the suspension liquid is decanted off, and the binder material is cured to affix the scintillator to the substrate. This is a general phosphor powder deposition process, providing the capability to deposit almost arbitrary types of scintillators.

Simultaneously with developing the capability to deposit phosphor spots, a laboratory apparatus was designed and fabricated to allow measurement of scintillator characteristics as a function of temperature under alpha particle irradiation. The central portion of the apparatus is contained within a vacuum chamber. A computer-controlled heater is mounted within the vacuum chamber and scintillator samples are placed on the heater surface. A collimated alpha source is pointed at the sample to produce scintillation pulses. The scintillation light is guided out of the chamber using a two-element lens system coupled to a chilled photo multiplier tube (PMT) with near single photon counting capability. The light is filtered before arriving at the PMT with a short wavelength-passing filter to remove as much as possible of the blackbody light emitted by the hot sample.

It has become apparent as scintillator materials were tested at progressively higher temperatures that blackbody emissions become the dominant source of light as compared to individual neutron induced pulses above approximately 400 °C. While this is advantageous for measuring temperature via multi-wavelength pyrometry, the intensity of blackbody emission in the visible bands at higher temperatures does mean that it will not be possible to operate the detector in pulse mode at higher temperatures. This is not believed to be particularly significant since current mode is available for high-flux, high-temperature situations. Figure 2 shows the developed scintillator testing apparatus in both a photograph and a schematic illustration.

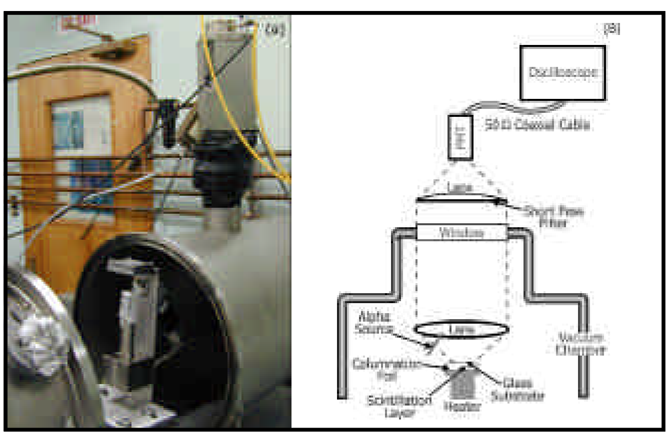



Figure 2. The laboratory scintillator testing apparatus appears both photographically (a) and schematically (b).

Significant progress has been made towards scintillation probe mechanical fabrication. Additionally, an aqueous technique to deposit silver films onto the interior surface of the light guides has been implemented, as has a slurry-based polishing technique to obtain a mirror finish in the light guides. While this technique is only applicable for light guides not exposed to temperatures above approximately 900°C due to silver vaporization, the technique is simple and requires no expensive equipment thus these light guides may be preferred in certain situations.

## Planned Activities

A platinum chemical vapor deposition process has been developed for higher temperature applications (limited by the fused silica maximum use temperatures of approximately 1,100°C). Thus far the process the process has only been demonstrated on short segments and researchers anticipate scaling the process as well as performing confirmatory survivability testing over the next two quarters.

Ohio State has made significant progress towards implementing the high temperature, near-core reactor testing environment and apparatus. Over the course of the next quarter, it is anticipated that nuclear testing will begin at OSU. The testing will include both lower flux and temperature testing in a subcritical pile; gamma only response testing using OSU's $^{60}$Co irradiator; reactor testing at ambient temperature; and finally, elevated temperature reactor testing.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Construction Cost Reductions in Generation IV Nuclear Energy System using Virtual Environments

Primary Investigator: Timothy Shaw, Pennsylvania State University

Collaborators: Panlyon Technologies; Westinghouse Electric Company; Burns and Roe Enterprises

Project Number: 01-069

Project Start Date: August 2001

Project End Date: September 2004

## Research Objectives

The objective of this project is to demonstrate the feasibility and effectiveness of using full-scale virtual reality simulation in the design of future nuclear power plants. Specifically, this project will test the suitability of Immersive Projection Display (IPD) technology to allow engineers to evaluate the potential cost reductions that can be realized in installation and construction sequences for Generation IV Nuclear Energy Systems. The intent is to see if this type of information technology can be used to improve arrangements and reduce both construction and maintenance costs, as has been done by building full-scale physical mock-ups for other systems.

## Research Progress

The contract was awarded in August 2001 and work began shortly thereafter. During a kickoff meeting, participants were introduced to the IPD technology. Potential uses as well as benefits and shortcomings of the technology were discussed. A number of test models were converted from existing virtual reality modeling language (VRML) files, downloaded from the Internet. This demonstration allowed half of the file conversion process to be tested. The test was expanded to include a number of different file formats available from the 3-D computer-aided design (CAD) package that Westinghouse used in the design of the AP600 plant. A method was developed for transferring and converting CAD files to a format that could be displayed in the IPD.

Once a suitable file conversion process was found, work began on developing the functionality of the software used to manipulate and interact with the virtual environment. Many features were added to the software over the course of the year. Several examples follow.

- A virtual measuring tape allows the user to quickly determine distances and clearances between objects.
- A virtual crane allows objects within the space to be moved in a manner similar to an actual crane.
- Components could be resolved separately and then tagged for identification, which allows the user to determine to which fluid system a piece of equipment belongs.
- Some of the valves have been modeled individually to allow them to be grabbed and moved or potentially operated.
- A system of hand gestures and voice-activated commands allows the user to point at components and identify them, toggle a measuring tape, operate a crane, or grab and move objects. Some of these interactions are depicted in Figure 1.

A simple collision detection scheme, which notifies the user if objects are touching, has been investigated for use in the installation sequence study and the maintenance evolution. Interaction with the environment is still somewhat awkward, but the software is being continually updated and refined. A configuration file allows the user to pre-program viewpoints and set the navigation speed.

Room 12306 within the Westinghouse Advanced Passive (AP 600/1000) Plant was chosen as a test bed for the virtual mock-up due to its relative complexity. The room is designed to contain five modules and piping assemblies, which are composed of piping from ten different fluid systems. The original files received from Westinghouse contained all of the geometry in a single VRML 1.0 file. The next iteration broke the room down by fluid system. The latest iteration has a large number of the valves modeled individually so they can be selected and manipulated. Other components are resolved by system. Methods for taking the existing geometry and increasing the resolution have been investigated so that

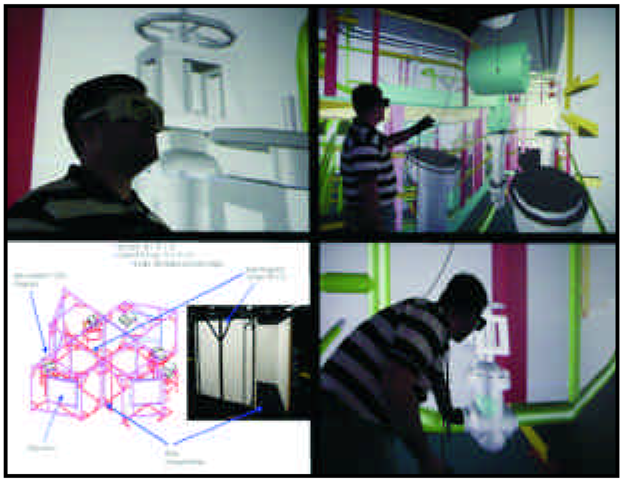


Figure 1: The graphics show features of the interactions possible using the virtual reality software being used to enhance construction studies for new nuclear power plants: (clockwise from upper left) Active Stereo Glasses, Virtual Crane, Interaction with Environment, and IPD Layout.

the individual pieces of piping that connect modules, called make-up or spool pieces, may be modeled.

During the initial development of the virtual mock-up, the room was reviewed by construction personnel to determine whether or not the technology provided sufficient realism, thereby providing a potential design tool. A survey was completed by the collaborators, which compared the virtual mock-up to computer models (3-D CAD) and physical mock-ups. Results of the surveys are currently being compiled.

In addition to the enhancement of room 12306, a lower fidelity virtual mock-up of the containment of the AP600 was constructed for educational and demonstration purposes. This model has been shown to industry personnel and government officials at the ICONE-10 and G-8 Energy Ministers' meetings. Response to the technology's potential has been very positive.

A number of tools have been developed to support the study of the installation sequence of the developed virtual mockup. Currently, the user is able to watch the 4D installation of prefabricated modules and makeup pieces. Different colored models are used to show the components before, during, and after installation. The software allows parallel activities to be displayed, as well. Installation sequences may be played back or viewed un-timed step-by-step within the virtual reality environment. Tools that assist the user in developing an installation sequence have been developed. A software "SELECT" function allows the user to change the color of models as they are installed in the space. Upon completion, the entire room appears in red. This information can be quickly loaded into the system, and the installation sequence can be played back for review.

## Planned Activities

Two installation sequence studies are being developed. The first study asks groups of construction management students to attempt to develop a construction sequence after a brief introduction to the components in the room. The students are given the freedom to change module boundaries and to cut pipes. The sequence they develop is then played back and evaluated. The second study asks experienced construction superintendents to develop a schedule using the isometric drawings provided by the designer. The schedule they develop will then be loaded into the virtual environment system for review and evaluation to identify constructability issues not revealed using the isometric drawings. The subjects will be surveyed concerning the benefits or drawbacks of developing the installation sequence with the assistance of the virtual environment technology.

NUCLEAR ENERGY RESEARCH INITIATIVE

## On-Line NDE for Advanced Reactor Designs

**Primary Investigator:** Norio Nakagawa, Ames Laboratory

**Collaborators:** Westinghouse Electric Company, LLC

**Project Number:** 01-076

**Project Start Date:** October 2001

**Project End Date:** September 2004

### Research Objectives

The extended refueling interval of Generation IV nuclear reactors creates new maintenance challenges. Current commercial reactors achieve high levels of availability and reliability by employing outage-based maintenance (i.e., performing methodical, periodic, off-line inspections, preventive maintenance, and component repair/replacement during planned refueling periods). Compared to the traditional 1- to 1.5-year refueling cycle, Generation IV reactors use extended refueling intervals such as 4 years and beyond. New approaches are required to keep maintenance from interrupting operation. The key strategy of this effort is to replace/augment current outage-based maintenance by on-line structural health monitoring, to ensure the current level of safety.

The project's specific objectives follow:

(1) Determine, based upon regulatory requirements and commercial reactor experience, the most appropriate inspection through a comprehensive review of each component of the Generation IV reactor design.

(2) Determine which inspections provide the greatest economic benefit when implemented as in-situ monitoring, in light of the inspection requirements and reduced off-line inspection opportunities.

(3) Optimize mechanical design parameters to simplify inspection.

(4) Develop the concept of a built-in structural integrity monitoring system using electromagnetic, ultrasonic, or radiation detectors, to be integrated into design for Generation IV nuclear power systems.

(5) Evaluate and characterize the performance of conceptual sensor systems by the use of physics-based simulation models.

(6) Enhance the capabilities of the simulation models to meet the challenges posed by unique power system environments.

(7) Select sensor types and materials, find their compatibility with hazardous environments, and examine their possible degradation.

Among various Generation IV reactor designs, the team pays particular attention to the International Reactor Innovative and Secure (IRIS) design. As with other Generation IV reactors, it has several general design goals: a) passive safety features; b) increased availability and economy; c) long-term, uninterrupted operation; d) environmental friendliness; and e) proliferation resistance. To meet these design requirements, most Generation IV reactors use compact, integrated designs, and are made compatible to operating with extended refueling cycles. IRIS, in particular, integrates the reactor core, steam generators (SG), and primary-coolant pumps in a single reactor pressure vessel (RPV). By design, the IRIS reactor is compact and cost-effective, and requires less maintenance because it has no large primary-water loop piping outside the RPV. In addition, there is less likelihood that the SG tubes will develop stress-corrosion cracking (SSC) since they operate in compression. However, the long refueling cycle (every four years for the IRIS-based design) poses a maintenance challenge because there will be fewer opportunities for periodic outage-based maintenance, as has been practiced for existing commercial reactors.

From the point of view of these key design features, future reactors such as IRIS call for a new maintenance strategy, i.e., not only relying on outage-based maintenance, but also actively performing on-line inspection and monitoring. The advantages of on-line health monitoring are multi-fold:

1) It is done while the reactor is in operation, not requiring shutdown for inspection/monitoring activities.

2) It is done remotely, thus greatly reducing exposure levels.

3) It allows investigators to perform continuous or on-demand system integrity verification, which means that deviation from normal operation can be detected in real time, maximizing potential options for issue resolution.

To demonstrate the concept, the project develops conceptual on-line sensor systems that will replace/augment outage-based maintenance.

## Research Progress

First, the IRIS design was reviewed to identify critical inspection needs. To date, several candidates for on-line monitoring have been identified: steam generators (SG), the reactor pressure vessel (RPV), reactor core, and coolant pumps. The potential issues identified for steam generators are magnetite deposit buildup, tube mount integrity, and tube integrity, while the concerns for the RPV and core are cracking and fuel activity anomaly, respectively. The coolant pumps may cause fatigue in their mounting mechanisms, while failure of a pump will create coolant flow anomaly inside RPV, leading to reactor instabilities.

Second, the team conceived on-line monitoring systems that can address identified monitoring needs, based on several candidate NDE methodologies. Specifically for SG, likely candidates are the eddy current (EC) method via fixed-site coils (Figure 1) and the ultrasonic guided-wave method via electromagnetic-acoustic transducers (EMAT) (Figure 2). For the RPV cracking and core anomaly monitoring, the uses of ultrasound testing (UT) via EMATs and radiation-based techniques based on silicon-carbide (SiC) detectors (Figure 3), respectively, will be explored. More recently, the above-mentioned pump issues were recognized, and the team has begun to explore the use of the use of hard-ray emission prompted by radioactive nitrogen-16 isotopes contained in the primary coolant.

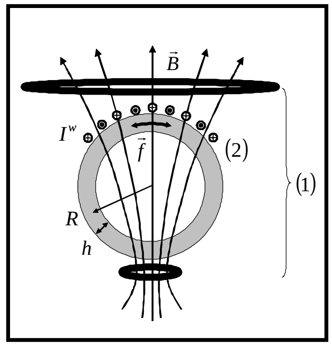



Figure 2. In this schematic of an EMAT system, the asymmetrical Helmholtz coil pair (1) will generate DC fields approximately perpendicular to the tube wall, suitable to generate torsional modes.

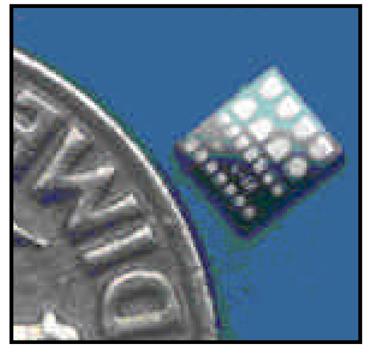


Figure 3. The photograph shows a patented radiation detector fabricated on a SiC semiconductor chip.specific design uses an encircling solenoid coil surrounding several tubes chosen by sampling.

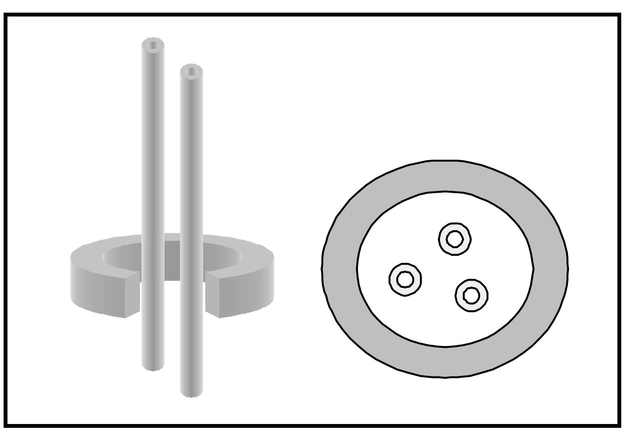


Figure 1. The diagram shows the conceptual encircling-coil design of an on-line EC inspection for magnetite deposit detection. A specific design uses an encircling solenoid coil surrounding several tubes chosen by sampling.

The third part of the team's activities has involved model-based studies of conceived on-line monitoring sensors to estimate their performance. In particular, EC sensor signals were estimated based on a simplified model. It was found that when the EC coils are operated at 20-30 kHz, they will produce distinctive magnetite-deposit signals, clearly standing out of the geometry-signal background. Guided-wave modes traveling along a tube have been worked out for the EMAT UT application. Among these, torsional modes are the primary candidates for use. A conceptual sensor system design was developed (Figure 2), in which the DC bias magnetic field distribution was actually computed to confirm the field lines drawn intuitively in Figure 2. A radiation propagation model based on transport equations is being developed for studying radiation-based techniques. The model involves both photon and charged-particle (electron and positron) sectors. The conceptual design of the algorithm has been completed.

## Planned Activities

These foregoing results and outputs indicate that the project is on schedule. Since no particular issues/concerns have been identified, the team plans to follow the original project schedule. The Year Two goals follow:

(1) Determine, by prediction, neutron and gamma-ray fluxes in critical areas under normal reactor operation.

(2) Complete integration of the charged-particle transport code into the photon transport code.

(3) Select a likely set of candidate on-line electromagnetic sensor designs according to the requirements of on-line SG tubing inspections.

(4) Complete the upgrade of EC model code for array probes.

(5) Determine the beam model for each of the EMAT designs.

In addition to these primary activities, the team will continue to interact with the IRIS design team members by participating in their team meetings, in order to mutually disseminate project developments. Exploration of additional on-line monitoring needs and methodologies will also be continued as further design developments of IRIS, and possibly of other Generation IV designs, become available.

Finally, public dissemination will continue of the team's on-line health-monitoring concept and its benefits when applied to nuclear power systems. The target audience includes broad technical communities, especially the NDE community.

# NUCLEAR ENERGY RESEARCH INITIATIVE

## Supercritical Water Nuclear Steam Supply System: Innovations in Materials, Neutronics, and Thermal-Hydraulics

Primary Investigator: M. Corradini, University of Wisconsin-Madison (UW)

Collaborators: Argonne National Laboratory (ANL)

Project Number: 01-091

Project Start Date: August 2001

Project End Date: September 2004

### Research Objectives

A nuclear reactor cooled by supercritical water[1] with a once-through direct power cycle would have a much higher thermal efficiency than a standard water reactor, and could be based on standardized water reactor components (light water or heavy water). The theoretical efficiency could be improved by more than 33 percent over that of other water reactors and could be simplified with higher reliability (e.g., a boiling water reactor without steam separators or dryers). Such improvements would be accompanied by a corresponding decrease in the nuclear plant levelized electricity cost, and thus, could make this nuclear steam supply system quite competitive in future electric power markets as a centralized power source. In addition, this concept would take full advantage of 50 years of light water reactor (LWR) technology, would allow for incremental as well as substantial improvements in reactor technology to maintain and enhance safety and reliability, and would provide flexibility in the fuel cycle to allow for substantial improvements in sustainability. This research project will make such a system technologically feasible by accomplishing the following objectives:

(1) Employ innovative ion implantation surface modification techniques to improve material compatibility at supercritical conditions. Plasma Source Ion Implantation techniques will be used to modify clad materials and demonstrate improved corrosion/wear resistance under supercritical thermal-hydraulic conditions.

(2) Use neutronics analyses to identify ranges of alternative fuel cycles, including variations in enrichment, refueling schedules, recycling, and conversion/breeding. These analyses would focus on coolant density effects at supercritical conditions to verify passive safety with comparisons from the standpoint of fuel burnup, flexibility, proliferation resistance as well as sustainable development, using quantitative metrics.

(3) Conduct thermal-hydraulic studies that focus on heat transfer and flow stability issues associated with coolant density changes for natural circulation of supercritical water. Scaled simulation experiments are to be designed and performed to provide heat transfer and stability data to be used in developing predictive tools.

[1] Tc=375°, Pc=220 ATM, Vapor Density and Liquid Density converge

### Research Progress

Work has gone forward on three tasks.

<u>Task 1. Cladding Materials, Surface Treatment, and Corrosion Loop:</u> Three cladding materials—Inconel 718, A1SI 316 austenitic stainless steel, and Zircaloy-2—were selected for this task. Samples of these materials have been obtained and surface preparation and coating were completed on several of the samples. Table 1 summarizes the three fundamentally different approaches of plasma surface modification that is underway.

**Table 1. Various Plasma-Based Surface Treatments**

| Plasma surface treatment | Improvements anticipated | Possible mechanisms |
|---|---|---|
| Ion implantation<br>- Room temperature*<br>- Elevated temperature | Hardness, wear, corrosion & oxidation resistance | Formation of hard compound phases, alteration of oxide nucleation mechanism and deeper modified layer (for elevated temperature) |
| Non-equilibrium surface alloying | Oxidation resistance | Formation of a more adherent oxide with slower growth kinetics |
| Heavy ion bombardment | Corrosion and oxidation resistance | Surface microstructural homogenization due to energetic ion-induced mixing |

*Underway this quarter

A supercritical water flow loop for corrosion studies has been designed and engineering work for construction has begun. Figure 1 shows the loop design along with the high heat flux heater design, which utilizes molten lead as the heat transfer media. The loop has an inner tube diameter of 4.29 cm and an outer tube diameter of 6.03 cm (Inconel 625).

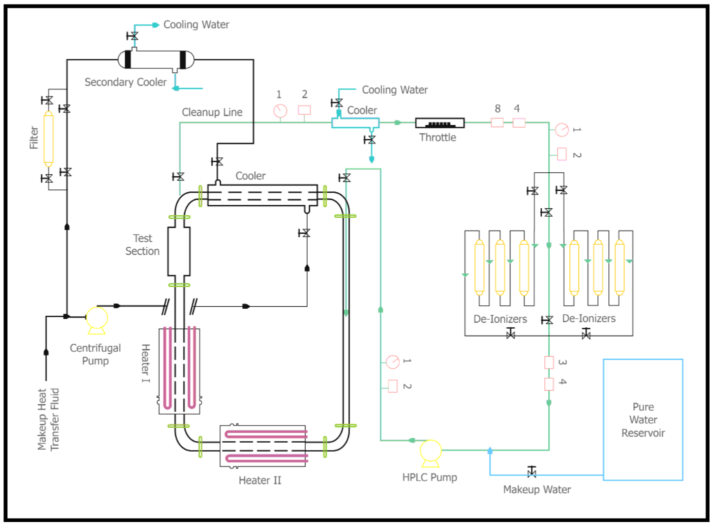



Figure 1. Flow Loop for SCW System

<u>Task 2. Neutronic Analysis of Coolant Density Effects and Cladding Materials:</u> The neutron physics of a supercritical water reactor (SCWR) is different from that of a standard (LWR) because of the low water density of supercritical water, high operating temperature of the SCWR, and the different cladding materials. In particular, the low density of supercritical water near the pseudo-critical temperature results in a harder spectrum in the SCWR related to a standard LWR. Although the WIMS8 code is well-developed for the LWRs the validation of the WIMS8 code is necessary because the neutron spectrum of the SCWR is different from that of the standard LWR. For the first stage of validation of WIMS8 code, the MCNP4C was used for benchmarking purposes.

The $k_\infty$ value and normalized pin power distributions of the 17x17 Westinghouse standard assembly, operating with a coolant/moderator density of 0.3 g/cm$^3$, predicted by MCNP4C and WIMS8, are provided in Table 2. The WIMS8 power distributions presented in this table were calculated with a 28-group transport solution. The maximum observed differences in $k_\infty$ are 387 pcm (0.3 percent). The normalized pin powers of the 28-group WIMS8 calculation are generally within c1c of the MCNP4C result and the root mean squares (RMS) of the power differences between WIMS8a and MCNP4C are very similar to the RMS of the statistical error of the MCNP4C.

**Table 2. Summary of the MCNP4C and WIMS8 Calculations**

| Calculation conditions | MCNP4C | | WIMS8 | | |
|---|---|---|---|---|---|
| | $k_\infty$ | RMS of MCNP power error [a] | Number of group [b] | Difference of $k_\infty$, pcm [c] | RMS of power error [d] |
| U enrichment=5.0%<br>Water density=0.3g/cm$^3$<br>Clad material=Zr-2<br>Temperature=300 K | 1.27325<br>c 0.00028 | c 0.44 | 6 | -211 | 0.47 |
| | | | 28 | -387 | 0.40 |
| | | | 172 | -318 | 0.40 |

a) Root mean square of the MCNP relative power error

b) Number of neutron energy groups in the transport solution

c) Difference of the $k_\infty = 10^5 * (k_\infty^{WIMS} - k_\infty^{MCNP})$, pcm

d) Root mean square of the difference power error between MCNP and WIMS8a

<u>Task 3. Natural Circulation Heat Transfer and Flow Stability Studies:</u> The design of a natural circulation loop of supercritical carbon dioxide is being finalized. The purpose of this test loop is to investigate natural circulation heat transfer and flow instabilities associated with the use of a supercritical fluid. Such instabilities are of interest because a number of proposed nuclear reactor systems with supercritical water would utilize the large density change across the pseudo-critical point to drive natural circulation cooling. Flow instability in the circulation may adversely impact the ability of the system to remove heat from the reactor core.

The design of the loop is shown schematically in Figure 2. It is a rectangular loop that consists of horizontal heating and cooling sections connected by two vertical pipes. The inner diameter and height of the loop are 0.014 meters and 2.0 meters, respectively. Calculations were performed for the loop operating at a constant pressure of 80 bar and a constant inlet temperature of 28°C. One of the operating curves produced by the calculations was the plot of the steady-state natural convection mass flow rate vs. the heater power. The plot showed that a peak in the mass flow rate occurs around the power needed to take the fluid through the pseudo-critical temperature along the heated section. Chatoorgoon recently proposed an ad hoc criterion postulating that this peak would correspond to an instability boundary [Int. J. Heat Mass Transfer, 44 (2001)]. This criterion suggests that a new type of flow instability, different from traditional instabilities, would arise in the region of negative slope on the plot of mass velocity vs. power. The natural circulation loop has been designed to investigate (and confirm or deny) this criterion. Specifications for the loop heating and cooling power as well as the dimensions (height and diameter) are consistent with this design requirement.

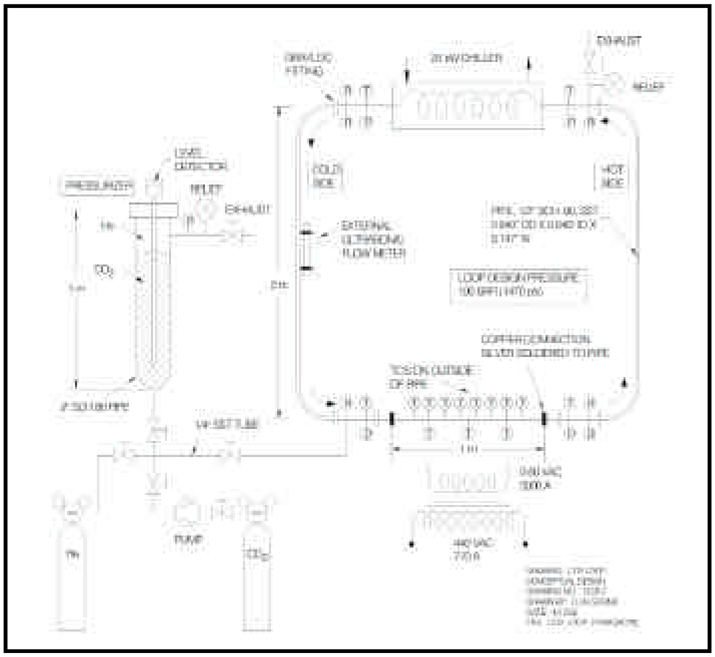



Figure 2. Supercritical $CO_2$ Natural Circulation Loop Design

## Planned Activities

Three future tasks have been planned.

Task 1. Initial tests of the surface-treated samples will be conducted to obtain a baseline analysis of the modified surface. These baseline tests will include potentiodynamic corrosion testing, wear, synergistic effects of wear and corrosion, and electron microscopy of the three reactor alloy materials being investigated in this project, before and after plasma surface treatments.

The supercritical water loop for corrosion studies will be constructed and become operational by the end of this calendar year. Once the loop is operational, the surface-treated samples will be tested in a prototypical supercritical water environment, and the test results will be compared to those of the baseline tests.

Task 2. The good agreement of WIMS8 with the MCNP4C calculations indicates that WIMS8 code is well-suited for predicting the eigenvalues and power distribution under the SCWR environment. However, since this benchmark has been done at the room temperature, the temperature effects on the cross-sections were not taken into account in this benchmark. Therefore, additional benchmark calculations will be performed after preparing the cross sections at the operating temperature.

Task 3. The supercritical $CO_2$ loop for studies of natural circulation heat transfer and flow stability will be constructed and become operational by the end of this calendar year. An experimental plan for proposed tests will be developed and issued. A safety plan for the experiments will be prepared to comply with the Argonne National Laboratory's environmental safety and health requirements.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Testing of Passive Safety System Performance for Higher Power Advanced Reactors

Primary Investigator: Jose N. Reyes, Oregon State University (OSU)

Project Number: 01-094

Project Start Date: August 2001

Project End Date: September 2004

## Research Objectives

The objective of this project is to assess the performance of various passive safety systems for advanced reactors operating at powers on the order of 3,000 MWth. The advantage of passive safety systems for core cooling following loss-of-coolant accidents is that they do not rely on safety grade pumps or alternating current power. Rather, they depend on natural driving forces such as gravity, compressed gas, and natural circulation to provide core cooling for an indefinite period of time after an accident. Currently, the AP600 is the only passively safe nuclear plant in the world that has received Design Certification from the U.S. Nuclear Regulatory Commission. Westinghouse has recently proposed the development of an AP1000 that would offer significant economic advantages over the AP600. Because of its multiple passive safety systems, the availability of a geometrically similar integral test facility at OSU, and the lower power AP600 database that can be used for purposes of comparison, the higher power AP1000 is an ideal candidate for this high power passive safety system study. The work will be carried out over a three-year period and includes a test facility scaling analysis, advanced plant experiment (APEX) facility modifications, and passive safety system tests and assessment for design basis loss-of-coolant accidents (LOCAs).

## Research Progress

Figure 1 is a flow chart describing the overall OSU research program. Significant progress was made during the first two quarters of this project. The first activity to be completed was a scaling assessment of the geometric, kinematic, and dynamic similarity between the APEX test facility components and operating conditions and those of the AP1000 reactor. As shown in Figure 2, APEX is a 1/4-length scale, integral system, test facility originally designed to simulate the important thermal-hydraulic behavior of the Westinghouse AP600. The scaling analysis for this project has identified the modifications that are needed to adequately simulate the new AP1000 reactor design concept using the existing OSU APEX test facility. The APEX facility modifications will be as follows:

- Increase core power from 600 kW to 1.0 MWth
- Replace the Pressurizer and Surge line
- Enlarge the Core Make-up Tanks (CMT)
- Enlarge Automatic Depressurization System (ADS) 4 valve flow area
- Reduce the CMT and Incontainment Refueling Water Storage Tank (IRWST) line resistances
- Upgrade the Data Acquisition System

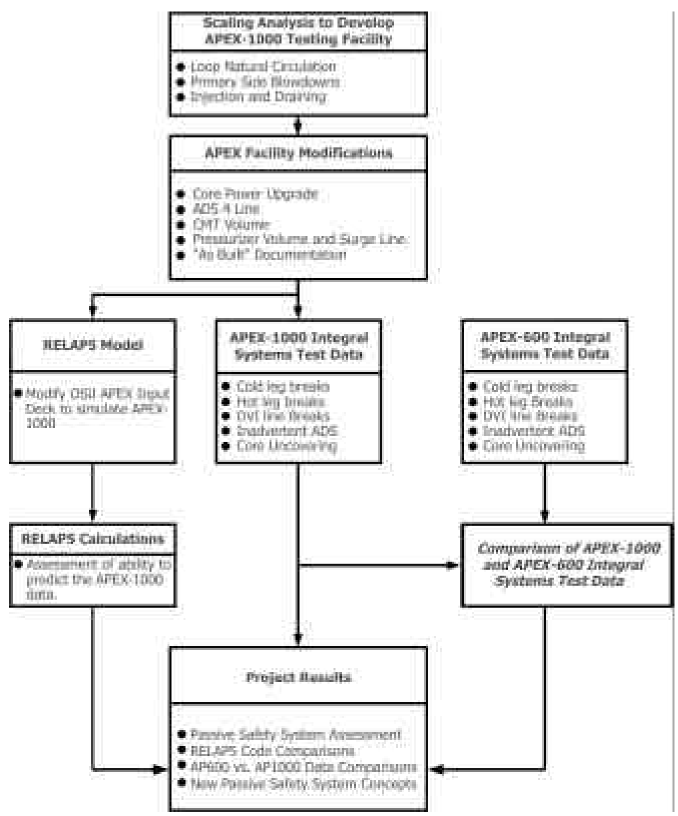



Figure 1. The flow chart shows the sequence of events in the OSU AP1000 Test and Analysis Program.

All of the system components will be constructed of stainless steel and will be capable of consistent operation at 400 psia while at saturation temperatures. All of the primary system components will be insulated to minimize heat loss.

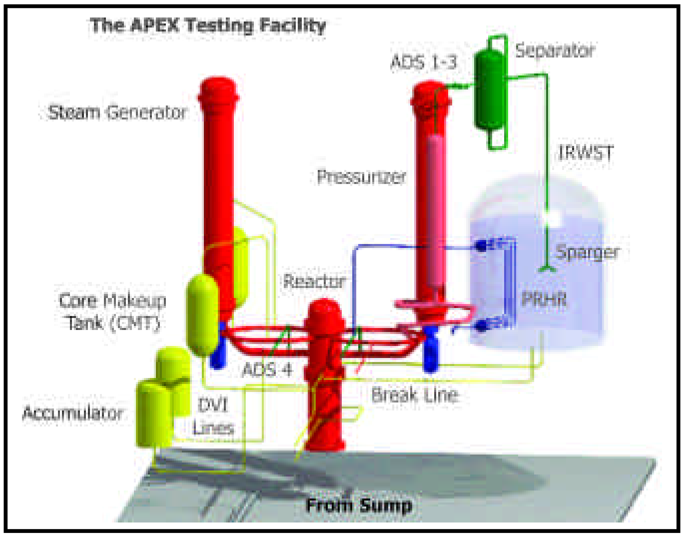



Figure 2. The figure is a graphical representation of the OSU APEX-1000 Testing Facility.

## Planned Activities

Having completed the scaling analysis, the key modifications will be made to the APEX Testing Facility and "as-built" drawings will be developed. All of the re-configured facility documentation will be labeled as APEX-1000. The facility modification details will also be incorporated into the APEX RELAP5 input deck in order to compare thermal hydraulic computer codes to the APEX-1000 test data. A variety of pre-test and post-test calculations will be performed.

After modifying the APEX Testing Facility, a wide range of integral systems tests will be performed. This includes cold leg breaks, hot leg breaks, inadvertent ADS operation, direct vessel injection (DVI) line breaks, and core uncovering tests. The APEX-1000 test results will be compared to the APEX-600 test results to gain an understanding of the effects of high power on passive safety system performance. All of the results will be documented in a final report. This includes a complete description of the test results, key comparisons to APEX-600 data, RELAP5 comparisons to key APEX-1000 data, and suggestions for new passive safety system concepts for next-generation, high-power reactors.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Engineering and Physics Optimization of Breed & Burn Fast Reactor Systems

Primary Investigator: Michael J. Driscoll, Massachusetts Institute of Technology

Collaborators: Idaho National Engineering & Environmental Laboratory; Argonne National Laboratory

Project Number: 02-005

Project Start Date: September 2002

Project End Date: September 2005

The term "breed & burn" (B&B) is used to refer to fast reactors in which reload fuel has a significantly lower enrichment than that required to sustain criticality. The deficit is made up by breeding new fissile material in the fresh fuel faster than its depletion in the older fuel in the core. In the ideal case, only depleted uranium make-up is required in the steady state, and no reprocessing is required. The most compelling attribute of B&B systems is their virtual guarantee of sustainability via significantly better utilization of uranium resources. The concept is found in the NERI field of endeavor F-1: "Nuclear Engineering-Advanced Nuclear Energy Systems," where Generation IV goals are addressed and summarized in Table 1. B&B is not an entirely new concept, although only a handful of investigators have published on this topic since it was first mentioned in 1958 by the Russian physicist, Feinberg. Moreover, the emphasis to date has been almost entirely on reactor physics. Complicated fuel shuffling schemes have been suggested, including temporary introduction of a moderator, and fuel in-core residence times in excess of 100 years (!) were put forward uncritically in this prior work.

The present proposal will determine the feasibility of achieving significant B&B benefits in practical plant designs. These will likely be based on gas coolant and carbide or metal fuel, to avoid introducing too much moderation, which spoils the ultra-hard spectra needed by this concept. High power density is also needed to accelerate fuel throughput and move quickly to an equilibrium fuel cycle. Overall project coordination will be provided by MIT; INEEL and ANL-WEST will contribute significantly in their areas of special expertise and in collaborative work on shared tasks.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Evaluation of Integral Pressurizers for Generation IV PWR Concepts

Primary Investigator: David K. Felde, Oak Ridge National Laboratory

Collaborators: Westinghouse Electric Company

Project Number: 02-018

Project Start Date: September 2002

Project End Date: September 2005

Integral pressurizers are a key design feature of several proposed pressurized water reactor (PWR) designs included in the Generation IV reactor concepts based on light water reactor (LWR) technology. These Integrated Primary System Reactor (IPSR) concepts are characterized by the inclusion of the entire primary system within a single pressure vessel, including the steam generators and pressurizer. For higher power output, forced circulation is required and designs include internal pumps, although relatively large natural-to-forced circulation flow ratios remain as favorable operating characteristics. As in conventional PWRs, the pressurizer and its interaction with the primary system are important factors in the dynamic control and stability of the reactor during both normal and off-normal conditions.

This project will develop the tools and methods needed to evaluate and characterize the functionality of integral pressurizer designs for new Generation IV reactor concepts. As part of this process, it is the intent to map the performance of integral pressurizers as a function of their design parameters (e.g., gas content, vapor volume, interface with the primary system). Based on the detailed analysis of pressurizer performance, the work will aim to propose pressurizer design solutions that will allow the reactor system to be simplified or withstand more severe accident scenarios.

The integral reactors are expected to have significant multi-dimensional effects because the entire primary circuit is accommodated within the reactor vessel. Analyses will be performed using a 3-D system thermal hydraulic code (RELAP5-3D) to characterize the inflow/outflow surge rates and the operating envelope required for the pressurizer of a generic integral reactor. As part of this process, safety objectives will be defined and incorporated into the models and applicable bounding conditions. Selection will be made of initiating events leading to the highest degree of reactor over-pressurization or de-pressurization. In parallel, characteristics of the supporting systems (e.g., spray, heaters) will be studied and operating regimes defined. The functional design requirements determined in this phase of the study will be used as input to more detailed analysis of specific integral pressurizer characteristics, as described below.

For each pressurizer type (e.g. steam, steam-gas), the design details that allow it to meet the functional requirements will be identified. The major issues affecting the design of an internal pressurizer for integral reactors are related to the behavior of the interface between the primary circuit and the pressurizer. For a steam pressurizer, the interface dictates the thermal stratification and thereby the separation of the steam phase from the generally cooler primary circuit flow. The steam-gas pressurizer relies on the addition of inert gas to the steam phase to reach the operating pressure conditions. The interface has to be designed properly to avoid large gas concentrations in the primary coolant. In order to examine the thermal and gas transport characteristics of the interface and to characterize the response of the pressurizer to system pressure changes, a comprehensive analysis using a Computational Fluid Dynamics (CFD) code will be made.

Complementary models, correlations, or calculational techniques will be applied, as necessary, to the base CFD code structure using the same solution algorithm. The following principal processes, occurring in the pressurizer, will be analyzed in order to evaluate a particular pressurizer design: expansion and associated compression of the gas phase; spray condensation (condensation on falling droplets); internal condensation and water flashing; condensate fall rate; bubble rise rate; diffusion controlled mass transfer on phase boundaries; gas solubility and content in water under different conditions; and so forth.

A small experiment will be undertaken to refine the CFD model and validate the code results. A scaled model of the pressurizer section of an integral reactor will be built. In-surge and out-surge flows will be determined on the basis of expected operational regimes developed previously. Steam and steam-gas combinations will be tested, varying the gas content. Different interface designs for the pressurizer will be investigated in order to minimize the mass diffusion and maximize the impulse transport through the interface.

Having established characteristics of the specific pressurizer types, an investigation will be made of factors that could potentially affect system stability, and in particular, their effect on the natural circulation capabilities of the reactor. For the gas-steam pressurizers in particular, the effects associated with the non-condensable gas content and its compressibility will be addressed. With information from the CFD studies, an effort will be made to evaluate the circulation stability of a generic natural circulation system typical of the integral PWR designs. The study will parametrically assess the effect of pressurizer parameters (e.g., gas content, gas volume, interface areas) on natural circulation stability.

The systematic approach developed in the previous phases of the study will be applied to a down-selected reactor/pressurizer design. Models and tools developed for the generic reactor will be applied to the specific design. Transient system code runs will be carried out to analyze the evolution of the same initiating events analyzed for the generic reactor for verification of results and of compliance with the established safety and design criteria. In addition, the stability tests and the analysis of the relationship between pressurizer parameters and natural circulation stability performed for the generic reactor will be repeated for the specific reactor/pressurizer design.

From the results of this study, it will be possible to develop a sound approach to pressurizer selection as a function of reactor characteristics, giving the designer the ability to make detailed design decisions. Critical design details and characteristics will be identified and guidelines for application of the results will be developed and provided in the final report.

The proposed project is a collaboration between ORNL and the Westinghouse Electric Company, with non-funded international support from the Politecnico di Milano (POLIMI), Italy, and the Comissao Nacional de Energia Nuclear (CNEN), Brazil.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Nuclear-Energy-Assisted Plasma Technology for Producing Hydrogen

Primary Investigator: Peter C. Kong, Idaho National Engineering and Environmental Laboratory

Project Number: 02-030

Project Start Date: September 2002

Project End Date: September 2005

Most of the energy currently used in the world comes from fossil energy sources. The world's supply of fossil energy is finite and presents a variety of environmental problems from mining and extraction activities to air pollution caused by emissions when they are burned. Although the world's store of fossil fuels has already diminished, the world's demand for energy will not diminish. Moreover, the developed world is less willing to tolerate environmental damage from future methods of energy production, and is actively seeking new solutions for its energy needs. Among all the alternative energy possibilities, hydrogen is the strongest candidate to meet the global demand for energy without sacrificing the environment-it does not emit any air pollutants. Since hydrogen's energy density is high, it is a highly efficient energy source that can be used for transportation, heating, and power generation. If it can be produced, transported, and stored economically and cleanly in large quantities, hydrogen can replace fossil fuels in

- Automobiles and other personal transportation,
- Industrial processes, and
- Distributed power applications.

Several technologies exist to produce hydrogen, but they have disadvantages. For example, fossil fuel reformers produce hydrogen from methane, gasoline, natural gas, or other fossil fuels. These reformer systems are complex and capital intensive and the hydrogen produced is of low purity. The technologies also create polluting emissions from the carbon, sulfur, and nitrogen compounds inherent in the fossil fuel. Additionally, hydrogen from reformers contains carbon monoxide, which requires separation to produce pure hydrogen. Hydrogen generated from fossil fuels must still be stored—either compressed in cylinders or liquefied and stored as a cryogenic liquid. Both of these storage mechanisms have limited consumer appeal, particularly for transportation and residential power applications.

Thermal cracking of fossil energy sources also produces hydrogen and solid carbon residue, but the processes require separation to obtain pure hydrogen. Electrolysis is also used to generate hydrogen from water. The water-electrolysis process consumes significant amount of electricity with low conversion efficiency, and is designed only for stationary use. Another option is the use of metal hydrides, but only for storing the energy that hydrogen produces. Metal hydride systems still require a source of hydrogen gas for producing hydrogen fuel. An improved process to produce hydrogen must be developed.

Sodium borohydride is a safe and concentrated hydrogen carrier compound and can store an impressive amount of hydrogen. For example, 1 liter of 44-weight percent sodium borohydride solution at 1 atmosphere can release about 130 grams of hydrogen. Sodium borohydride releases more hydrogen and has a higher density of hydrogen than other sources of hydrogen. For example, cryogenic liquefied hydrogen has a density of 70 gm/lit. Hydrogen pressurized to 6,000 psi has a density of only 36 gm/lit. Rare-earth-nickel alloys can store hydrogen up to a density slightly higher than liquid hydrogen but still quite a bit less than that of sodium borohydride. However, the alloy is very expensive and not as easily handled as a liquid. The borohydride solution is also much easier and safer to handle than liquid or high-pressure hydrogen. The current gasoline-distribution infrastructure for automobiles can be easily converted to dispense "sodium borohydride fuel" for vehicles.

Sodium borohydride can be produced from sodium borate although at present, no technology exists to do so economically. Development of a nuclear-power-assisted plasma technology is proposed for economically mass-producing sodium borohydride from sodium borate.

A successful nuclear-power-assisted plasma technology to convert sodium borate to sodium borohydride will have a long-term significant economical benefit to the nuclear power industry. During peak operation, nuclear power reactors will generate electricity to meet peak commercial demand, and during off-peak operation, the nuclear reactor will supply electricity and nuclear process heat to produce sodium borohydride. Producing sodium borohydride during off-peak hours will in turn increase the demand for the nuclear industry.

We have assembled a team of highly experienced and qualified researchers to develop a new nuclear-energy-assisted plasma technology to produce hydrogen. Our proposed technology does not have the disadvantages of existing hydrogen-producing technologies. In contrast to the existing hydrogen-producing technologies, the proposed process to mass-produce sodium borohydride from sodium borate is

- Efficient,
- Economical,
- Environmentally acceptable, and
- Safe.

It will also help the country convert to a hydrogen economy. Sodium borohydride solution is also much easier and safer to handle than liquid or high-pressure hydrogen. This technology will help in facilitating hydrogen as the energy source to power the world.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Coupling of High Temperature, Lead-Cooled, Closed Fuel Cycle Fast Reactors to Advanced Energy Converters

Primary Investigator: James J. Sienicki, Argonne National Laboratory (ANL)

Collaborators: Oregon State University; Forschungszentrum Karlsruhe

Project Number: 02-065

Project Start Date: September 2002

Project End Date: September 2005

The three-year program of research and development aims to develop high-temperature modular nuclear plant concepts that take advantage of the sustainability benefits of a fast neutron spectrum core; the safety benefits of molten lead primary coolant; and the cost advantages of modular construction, factory fabrication, and simplification with natural circulation heat transport. At the same time these plant concepts would achieve sufficiently high coolant temperatures to drive an advanced power conversion system-for example, a gas turbine Brayton cycle using supercritical carbon dioxide-providing efficiencies competitive with those claimed for high temperature gas reactor (HTGR) concepts. Unlike HTGRs, the concepts provide the sustainability and economic fuel cycle benefits of a liquid metal-cooled fast reactor. Unlike conventional fast reactors, utilization of a gas turbine, compressors, recuperator, precooler, intercoolers, and supporting components offers radical plant simplification, reduced staffing levels, and cost savings, as well as greater efficiency relative to a Rankine cycle water-steam system, but at traditional liquid metal reactor (LMR) temperatures of approximately 550°C.

Lead ($T_{mp}$ = 327°C) is selected as the primary coolant based upon its high boiling temperature (1,740°C) and inertness; lead does not burn when exposed to air. When operating at higher-temperature, Brayton-cycle conditions, the high melting point (327ºC) ceases to be as large a problem as under Rankine cycle conditions. Lead is less corrosive than bismuth, especially at elevated temperatures. Small module power (e.g., approximately 400 MWth) enables 100+ percent natural circulation of the primary coolant, enhancing plant simplification, reliability, cost savings, and passive safety. The fast spectrum core with negative reactivity feedbacks facilitates nearly autonomous operation whereby the core power automatically adjusts itself to load changes as a result of inherent physical processes. Heat rejection to a gas also favors autonomous load following over a wider range of power levels. The reactivity feedback coefficients together with a passive reactor exterior cooling system utilizing air driven by natural circulation also effect passive core power shutdown in the event of accidents such as a loss-of-heat sink.

The utilization of supercritical carbon dioxide as the Brayton cycle working fluid could provide cycle efficiencies of about 45 percent at core outlet temperatures as low as 550°C. Increases in efficiency to well over 50 percent could be achieved with supercritical carbon dioxide by increasing the lead temperature. The achievement of such high efficiencies, even at traditional fast reactor temperatures, is a result of the low amount of work required to compress carbon dioxide immediately above the critical pressure as compared to the case of nonsupercritical He or $CO_2$. The supercritical $CO_2$ approach is particularly attractive because it works at temperatures traditionally reached in LMR systems, whereas if gaseous (i.e., nonsupercritical) helium or carbon dioxide were utilized as the Brayton cycle working fluid, cycle efficiencies of 45 to 50 percent would be achieved only by heating the gas to a core outlet temperature of nearly 900°C. In that case, a main challenge would be the identification of cladding, fuel, and structural materials for use with molten lead at elevated temperatures as well as innovative techniques for the manufacture of components from these materials. The need for experiemental data to undertake further development will be evaluated.

The goal to move away from the Rankine steam cycle to modern energy converters will not stop at the Brayton cycle. As an alternate to a gas-turbine energy converter, magnetohydrodynamic (MHD) generators for direct power conversion can potentially take advantage of the high electrical conductivity of liquid metals such as molten lead. A MHD generator converts fluid kinetic energy to electrical energy.

Argonne National Laboratory, as well as the Forschungszentrum Karlsruhe in Karlsruhe, Germany, will partner with Oregon State University (OSU) on various aspects of the project.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Experimental Verification of Magnetic Insulation for Direct Energy Conversion Fission Reactors

Primary Investigator: Donald B. King, Sandia National Laboratories

Collaborators: Texas A&M University; General Atomics

Project Number: 02-068

Project Start Date: September 2002

Project End Date: September 2005

This project will investigate the feasibility of direct energy conversion by designing, building, and testing prototype cells with advanced magnetic isolation and insulation technologies.

This research will establish the feasibility of developing reactors that directly capture the energy of nuclear fission fragments to produce electricity. With no intermediate conversion to thermal energy, the efficiencies of such reactors are not subject to classical thermodynamic limitations. The potential maximum efficiency of a direct energy conversion reactor is approximately 50 percent and is independent of temperature. As high temperatures and pressures are not required, large safety margins and passively safe design should be achievable. These advantages, combined with integral power conversion and modular design, present an opportunity to develop a low-cost reactor system.

Concepts to achieve direct energy conversion of fission fragments were investigated during the 1950s and 1960s. Experiments demonstrated the basic physics of the concept, but technical shortfalls prevented the attainment of operational goals. Magnetic solenoids required to capture the fission fragments and insulators to withstand high voltage gradients due to capture did not exist. Dramatic improvements in relevant technological disciplines have occurred since this time, including magnetic and insulation development, and are directly applicable to this direct energy conversion project.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Innovative Low-Cost Approaches to Automating QA/QC of Fuel Particle Production Using On-line Nondestructive Methods for Higher Reliability

Primary Investigator: Ronald L. Hockey, Pacific Northwest National Laboratory

Collaborators: General Atomics; Iowa State University; Oak Ridge National Laboratory

Project Number: 02-103

Project Start Date: September 2002

Project End Date: September 2005

Sub-millimeter TRISO fuel particles having multiple layers of pyrocarbon and silicon carbide are used in several current research systems and proposed advanced nuclear reactor fuel designs. The performance of these micro-spheres is a key component in system containment and depends particularly heavily on the properties and performance of the silicon carbide layer. Present day quality assurance and quality control (QA/QC) methods, done manually and in many cases destructively, are unable to test economically the large numbers of fuel particles that are required in fuel fabrication for advanced reactor concepts.

The project is designed to provide the United States with key enabling advanced inspection technologies for application to "fuel particles" (TRISO particles). These technologies are required for the economical production of reactor fuels being proposed for several Nuclear Power 2010 and Generation IV designs. The project will explore, adapt, develop, and demonstrate innovative nondestructive test methods that will provide in-line measurements for qualification of multilayered (TRISO) nuclear fuel particles and provide improved QA/QC.

The project will focus on nondestructive technologies that can be automated for production speeds, particularly those with potential for implementation as in-line measurements. Supporting studies will be performed on techniques with potential for either on-process implementation (where average properties of a batch of particles can be characterized) and those that can be used to give enhanced off-line measurements. The primary task for both the in-line and off-line tests will be to provide standard signatures for both acceptable particles and the most problematic types of defects. The data from the signatures will be used as the basis for establishing and demonstrating a multiple attribute "Quality Index," which can be used to integrate data and can be applied to grade both individual and batches of particles.

The primary thrust of the project will be in-line measurements, and this will focus on the assessment of the potential of electrical property measurements, which have potential for noncontact, rapid, volumetric property determination. It is proposed that these will be combined with advanced optical measurements to give shape and size assessment. It is intended that the data from these two methodologies will be integrated to give a Quality Index for both individual particles and batches of particles.

Supporting studies and particle characterization will be performed using high resolution computed tomography, acoustic microscopy, and resonance ultrasonic spectroscopy. Data from these studies will be used to assess the performance of the integrated electrical and optical measurements. If required to provide additional in-line characterization, an additional technology, from among those identified above, will be developed to support the optical and electrical measurements. The potential for the use of low-frequency ultrasonics as an on-process tool for monitoring batch properties will be evaluated.

The benefits from the successful completion of the project follow:

- Demonstration of the feasibility of using electrical measurements (eddy current/dielectric constant), integrated with advanced optical testing, for on-line TRISO QA/QC
- Development and demonstration of a Quality Index to measure both individual and batch conformity
- Sets of well-characterized surrogate particles for use in QA/QC technology evaluations

- An assessment of the capabilities of using acoustic microscopy, resonant ultrasound spectroscopy (RUS) and high resolution computed tomography for both in-line and/or advanced off-line NDE/QA-QC measurements on TRISO fuel particles
- Provision of proof-of-principle data for the use of transmission and diffuse field ultrasound for on-process monitoring and to provide improved quality control
- Availability of a family of QA/QC tools, which have been evaluated on surrogate fuel particles and are ready to be transitioned for use on a pilot plant scale for a TRISO fuel, or similar nuclear fuel fabrication line

NUCLEAR ENERGY RESEARCH INITIATIVE

## Model Based Transient Control and Component Degradation Monitoring in Generation IV Nuclear Power Plants

Primary Investigator: James Holloway, University of Michigan

Collaborators: Westinghouse Electric Company; Sandia National Laboratory; Dominion Generation

Project Number: 02-113

Project Start Date: September 2002

Project End Date: September 2005

A project is proposed to support the development of advanced nuclear power technology and to help position it as a highly competitive and safe method of energy generation. The project will develop a highly advanced and integrated methodology for constructing model-based control systems for Generation IV-based nuclear generating stations. The project will also develop an advanced approach for monitoring nuclear plant systems for system degradations. These two tasks are united by their reliance on smart sensor networks that map sensor signals to plant state information. This plant state information is used to connect models of plant state to the actual plant state. Nonlinear state-space control algorithms based on a Hamiltonian formulation of the control problem can then provide robust and automatic plant control in a wide variety of plant transient maneuvers, such as start-up, shutdown, and load follow maneuvers, including large or total load rejections. By providing smooth transient control without reactor trip these control systems can greatly improve both plant safety and economics. The quest for long-life cores in highly integrated and modular reactor designs places great demands on the already difficult maintenance systems of nuclear power stations. Development is proposed of a systematic statistical methodology for monitoring plant performance degradation. By solving a Master equation for the probability of finding the plant in a given system state and having a given set of component states, it is possible to determine the probability that the plant is in a given component state, given a set of plant sensor signals. Such advanced degradation monitoring will allow nuclear plant operators to optimize plant maintenance procedures that are subject to both safety and economic factors.

The work proposed will provide the nuclear engineering community with two new capabilities:

(1) A method to develop robust nonlinear control algorithms that combine plant sensor measurements with a physical model of key plant systems.

(2) A methodology for plant system degradation monitoring based on comparing plant sensor readings with a physical model of key plant systems.

These methods for fusing sensor data with physical models of plant systems will allow nuclear plant engineers to design optimal maintenance and control strategies at the onset for the new generation of nuclear plants. They will provide nuclear plant operators with tools to operate their plants safely and efficiently within the complex energy market of the 21st century.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Centralized Hydrogen Production from Nuclear Power: Infrastructure Analysis and Test-Case Design Study

Primary Investigator: William A. Summers, Savannah River Technology Center

Collaborators: General Atomics; University of South Carolina; Entergy Nuclear, Inc.

Project Number: 02-160

Project Start Date: September 2002

Project End Date: September 2005

The National Energy Plan proposed by President Bush foresees that energy from hydrogen will have an increasing role in the national economy, and that a large-scale, hydrogen-based, energy economy will augment the current fossil fuel energy economy, reducing the nation's dependence on imported petroleum.

Achieving the vision of a hydrogen-based economy requires safe, cost-effective methods of producing and distributing hydrogen in the quantities needed to support a major part of the Nation's energy and transportation needs. Also required is a strategy for making the transition to such an economy, especially because the infrastructure needs for centralized hydrogen production are considerably more complex than those for the alternative method of distributed hydrogen production using electrolysis.

The objective of this research is to identify, characterize, and evaluate the critical technical and economic issues associated with a new and innovative approach to centralized hydrogen production-thermochemical decomposition of water using heat from a nuclear reactor. These issues include hydrogen production, storage, distribution, and end-user integration.

Outcomes of this research will include the information needed to evaluate the technical feasibility and economic attractiveness of nuclear reactor-produced hydrogen, as well as the detailed characteristics for a commercial prototype system and an analysis of the economics of building such a system. The resulting methodology will help in developing the actual production facilities and related infrastructure. Technology gaps identified in this study will be the basis for future research projects designed to overcome barriers to implementation.

This research will define the process and infrastructure needed for nuclear hydrogen production to become a reality. In the process, the project will take advantage of many past and current studies of either production or infrastructure issues. However, although hydrogen infrastructure studies have been performed, no comprehensive analysis exists of an integrated nuclear reactor-thermochemical production system and the required supporting infrastructure. Therefore, this study will be unique in examining the integration of nuclear and thermochemical processes with infrastructure, and will define the engineering and economic factors needed to deliver nuclear-produced hydrogen to end users.
This study will build on existing design studies being supported by the Department of Energy through the Nuclear Energy Research Initiative, and will have two phases:

- Phase A, Infrastructure Analysis-conduct a comprehensive examination of nuclear hydrogen production methods and related hydrogen infrastructure systems. From this phase will come the detailed information needed to evaluate the technical feasibility and economic attractiveness of nuclear-thermochemical hydrogen production and to build a prototype commercial system.
- Phase B, Test Case Preconceptual Design-develop more specific, detailed results by conducting a test-case design study and economic analysis that hypothesizes thermochemical hydrogen production at a specific site and with a specific end-user. The Department of Energy's Savannah River Site and an existing local chemical plant will be the basis for this study. This phase is particularly applicable to determining the feasibility of using nuclear reactor-produced hydrogen for industrial applications such as oil refineries and chemical plants during transition to a large-scale, hydrogen-based energy economy.

The proposed project is led by the Westinghouse Savannah River Company (WSRC) through its applied research and development laboratory, Savannah River Technology Center (SRTC). Supporting SRTC will be a highly qualified team of two major industrial partners, General Atomics (GA) and Entergy Nuclear, Inc.; a leading university partner, the University of South Carolina (USC); and two experienced hydrogen consultants, Mr. Robert B. Moore, (retired, Air Products and Chemicals), and Dr. Joan Ogden, Princeton University. The team members will contribute extensive expertise and experience in every discipline required to make this project a success.

WSRC, with over 50 years of nuclear and hydrogen expertise as well as extensive research and development (R&D), production, and project experience, will take the lead in defining the hydrogen storage, transmission, and delivery systems, and will provide overall project management. GA brings a wealth of experience in nuclear reactor design and the thermochemical process, and will lead the process definition in these two areas. Entergy Nuclear, the nation's second largest nuclear power plant operator, will validate the preliminary design and cost information and provide an overall utility-company perspective on the project. USC has considerable expertise in hydrogen and fuel cell technology, and will lead the effort to develop an economic model to evaluate the various hydrogen infrastructure scenarios. USC will be supported in their tasks by the two key consultants, Robert Moore and Dr. Joan Ogden, both of whom have extensive backgrounds in hydrogen infrastructure studies and planning.

Hydrogen produced from nuclear power not only has many attractive environmental advantages, including the reduced emissions of nitrous oxides, sulfur, and global warming gases. It also has the potential to impact the Nation's energy security by reducing a dependence on imported oil. However, uncertainty about the supply system and the price of end-use hydrogen precludes an accurate assessment of hydrogen's potential future contribution to the national energy supply. This unique, comprehensive study will help define that future by providing valuable information for assessing the merits of nuclear hydrogen production, both for near-term hydrogen supply for chemical plants and for longer-term hydrogen supply for the hydrogen economy.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Near-Core and In-Core Neutron Radiation Monitors for Real Time Neutron Flux Monitoring and Reactor Power Level Measurements

Primary Investigator: Douglas S. McGregor, Kansas State University

Project Number: 02-174

Project Start Date: September 2002

Project End Date: September 2005

There is a need for neutron radiation detectors capable of withstanding intense radiation fields, of performing "near-core" reactor measurements, of pulse mode and current mode operation, and of discriminating neutron signals from background gamma ray signals. The detectors should also be tiny enough to be inserted directly into a nuclear reactor without significantly reducing or altering the neutron flux. Such devices can be used to monitor nuclear reactor power levels in "real-time."

A method is proposed here to accomplish these requirements with a new type of compact neutron detector fabricated through the utilization of present day micro-machining technology. The basic device consists of a miniaturized gas-filled chamber with either $^{10}B$ or $^{235}U$ inside coatings. The device width can be reduced to 1 mm or less while retaining up to 7 percent thermal neutron detection efficiency. The device is extremely radiation-hard and should continue to operate after exposure to neutron fluences exceeding $10^{16}$ n/cm$^2$. Furthermore, the compact design reduces background gamma ray interference. The device can be manufactured from a variety of materials, including common semiconductor and insulating materials. Overall, the device will be inexpensive to reproduce and operate.

The compact devices will be deployed in and around the KSU TRIGA reactor and tested as real-time neutron flux and power monitors. Inversion models will be developed to correlate the detector measurements with reactor power levels and performance.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Development of a Supercritical Carbon Dioxide Brayton Cycle: Improving PBR Efficiency and Testing Material Compatibility

Primary Investigator: Chang H. Oh, Idaho National Engineering and Environmental Laboratory (INEEL)

Project Number: 02-190

Project Start Date: September 2002

Project End Date: September 2005

Generation IV reactors will need to be intrinsically safe, having a proliferation-resistant fuel cycle and several advantages relative to existing light water reactor (LWR) systems. They, however, must still overcome certain technical issues and the cost barrier before it can be built anywhere in the world. South Africa wants to build the German-type PBR, but it has no detailed calculations on postulated accidents using the German design, there are unresolved technical issues, and the cost factor is still unfavorable.

A fundamental study is proposed to determine how to make the gas-cooled reactors safer and more economical, to meet the world's power requirements for the next generation.

The proposed project establishes a nuclear power cost goal of 3.3 cents/kWh in order to compete with fossil combined-cycle, gas-turbine power generation. This goal requires approximately a 30 percent reduction in power cost for state-of-the-art nuclear plants. It has been demonstrated that this large cost differential can be overcome only by technology improvements that lead to a combination of better efficiency and more compatible reactor materials. The proposal outlines

(1) development of a supercritical carbon dioxide Brayton cycle,

(2) improvement of the plant net efficiency by using the supercritical carbon dioxide Brayton cycle, and

(3) testing of material compatibility at supercritical conditions and high temperatures.

<u>Developing a Supercritical Carbon Dioxide Brayton Cycle and Improving Efficiency:</u> Supercritical carbon dioxide (SC $CO_2$) has a moderate critical constant, $T_c$ = 31°C and $P_c$ = 7.29 MPa, and has a unique heat transfer capacity at the supercritical condition. For example, the density of SC $CO_2$ is higher than helium by a factor 2000 and higher than supercritical water (SCW) by a factor of 1.5 to 2 at a temperature of 920°C and above. SC $CO_2$ has advantages over even SCW and helium in terms of heat transfer and higher molecular weight. The heat capacity term (mass flow rate times heat capacity) is higher than that for helium because of higher density. SCW has a much higher pressure (22 MPa) at the critical condition and a very narrow range of high heat capacity around the critical temperature of 374°C.

INEEL calculations for the Brayton cycle indicate that SC $CO_2$ has a 55 percent cycle efficiency versus 41 percent for helium for the reference PBR design of INEEL and MIT. The higher efficiency is achieved at a lower turbine inlet temperature for SC $CO_2$, 535.4°C versus 850°C for helium. The higher molecular weight results in less work in compression, which contributes to a higher efficiency for the SC $CO_2$ Brayton cycle.

The use of SC $CO_2$ as a coolant in the secondary PBR is very attractive because the core outlet temperature can be increased, which will increase the plant net efficiency by more than 60 percent.

<u>Testing Material Compatibility:</u> It is proposed to characterize the creep deformation of Inconel MA 754 and 758 over a range of temperatures from 850°C to 1050°C and stresses within the power law creep regime. By varying the temperature at constant stress and the stress at constant temperature, it will be possible to determine the numerical values of the activation energy and power law exponent for creep, and whether a threshold stress formalism applies for these materials or if the ARZT type model is more appropriate. This characterization will allow the proper constitutive equation to be determined, so that the deformation behavior can be calculated for a long service time in the temperature and stresses expected for the advanced reactor concept described above.

In addition to suitable mechanical properties, the alloys must resist environmental degradation for extended periods of time for the conditions expected in this reactor concept.  Preliminary analysis suggests that the nickel-based alloys with 20 to 30 percent Cr content will exhibit reasonable resistance to degradation by supercritical $CO_2$. An investigation is proposed of the interaction of MA 754 and 758 in supercritical $CO_2$ using thermogravimetric analysis combined with surface analysis to examine the possible chemical interaction mechanism(s) (e.g., breakdown of the passivating Cr oxide or carburization), at temperatures and pressures of interest.

# NUCLEAR ENERGY RESEARCH INITIATIVE

## Hydrogen Production Plant Using the Modular Helium Reactor

Primary Investigator: Arkal Shenoy, General Atomics

Collaborators: Idaho National Engineering & Environmental Laboratory; Entergy Nuclear Inc.; Texas A&M University

Project Number: 02-196

Project Start Date: September 2002

Project End Date: September 2005

There is a large and growing demand for hydrogen both in the United States and worldwide, with the bulk of the hydrogen being produced by steam reforming of methane. Hydrogen, along with electricity, are expected to dominate the world energy system in the long term. As the United States and the rest of the world transitions to a hydrogen economy, hydrogen will be used increasingly by the transportation, residential, industrial, and commercial sectors of the energy market. Eventually, an alternative source of hydrogen will be needed because

(1) the demand for natural gas is outpacing its production, and

(2) steam reforming of natural gas is not environmentally friendly because it produces the greenhouse gas $CO_2$.

A promising alternative source of hydrogen is to use process heat from a high-temperature nuclear reactor to drive a set of chemical reactions that produce hydrogen. Preliminary evaluations have shown that the sulfur-iodine (SI) process can produce hydrogen with high efficiency when driven by the 850°C to 950°C process heat from a Modular Helium Reactor (MHR). The SI process produces highly pure $H_2$ and $O_2$, with formation, decomposition, regeneration, and recycle of the reagents $H_2SO_4$ and HI. Preliminary economic assessments have shown that an MHR-driven SI plant can produce hydrogen economically, especially if the cost of natural gas increases because of increased demand. The MHR's high-temperature capability, advanced stage of development relative to other high-temperature reactor concepts, and passive-safety features make it ideally suited as the heat source for producing hydrogen. The work proposed here is the next logical step-to develop a conceptual design for a hydrogen production plant that integrates an MHR reactor system with an SI-cycle hydrogen production plant. Figure 1 shows an artist's conception of the integrated plant, referred to as the $H_2$-MHR. As an added measure of safety, the reactor system is located below grade and isolated from the $H_2$ production system through the use of intermediate heat exchangers. The $H_2$-MHR represents a significant advancement of nuclear technology and offers a safe and potentially economical source of clean, renewable hydrogen.

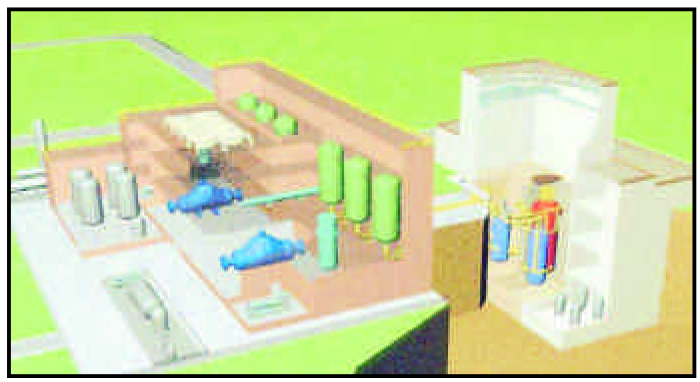


Figure 1. The schematic of the $H_2$-MHR plant shows the modular helium reactor integrated with an $H_2$ production plant (source: Generation IV Higher Temperature Reactor Materials Workshop, La Jolla, CA, March 18, 2002; figure prepared by Japan Atomic Energy Research Institute).

The proposed project will span a three-year period. During the first six months of the project, a systems-engineering approach will be used to prepare a Plant Functions and Requirements document. This document will provide the basis for developing the conceptual design of the $H_2$-MHR plant. Annual reports will be issued at the end of Project Years 1 and 2 to document the work performed during these years. An $H_2$-MHR Conceptual Design Report will be the final deliverable, to be issued at the end of Project Year 3. The work proposed here supports all of the NERI program objectives and will provide the Department of Energy, utilities, and energy-policy planners with precisely the type of information needed to make decisions regarding additional research and development for producing hydrogen using nuclear energy.

A team consisting of General Atomics (GA), Idaho National Engineering and Environmental Laboratory (INEEL), Entergy, and Texas A&M has been assembled to perform the proposed work. GA will be the lead organization and will be responsible for project management, plant definition, reactor system design, and plant integration. Texas A&M will have lead responsibility for developing the hydrogen production system design. INEEL will have lead responsibility for performing plant assessments, trade studies, and sensitivity analyses. Entergy, a major nuclear utility with a strong interest in hydrogen production, will function as a non-funded participant and will periodically review the design work from the perspective of a potential customer. Each organization is highly qualified and highly motivated to work on this project.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Nuclear Reactor Power Monitoring Using Silicon Carbide Semiconductor Radiation Detectors

Primary Investigator: Don Miller, Ohio State University

Collaborators: Westinghouse Savannah River Company; General Atomics

Project Number: 02-207

Project Start Date: September 2002

Project End Date: September 2005

This proposal is directed to the design, development, and assessment of a new paradigm in ex-core neutron flux monitoring for nuclear power plants. The proposed system is based on the use of silicon carbide (SiC) neutron sensors configured in arrays, technology that has been under development by Westinghouse since 1994. There are two fundamental characteristics of such arrays that distinguish them from current technology, which employs a variety of gas-filled neutron sensors:

(1) They operate in pulse mode over a wide dynamic range, which permits pulse spectroscopy, and

(2) they are relatively small in physical size, which permits measurements at discrete physical locations.

To access these characteristics, a collaborative program among the Ohio State University (OSU), Westinghouse, and General Atomics (GA) is proposed that will investigate the use of SiC-based sensor arrays as ex-core neutron monitors in the International Reactor Innovative and Secure Reactor (IRIS), which is being developed by Westinghouse Electric Company and in the prismatic-core, gas turbine modular helium-cooled reactor (GT-MHR), which is being developed by General Atomics.

The proposed three year research program will identify advantages and disadvantages associated with the use of SiC neutron sensors, examine solutions for overcoming difficulties associated with their use and will develop and evaluate methods for improving the performance of SiC based neutron sensor channels.

The following deliverables are identified as key products of the proposed research program.

(1) Selection of the optimum locations for SiC-based neutron power monitors in both the IRIS and the GT-MHR. Factors that will be considered include the power monitoring requirements as well as expected detector sensitivity and presence of gamma ray background.

(2) Evaluation of other applications and opportunities offered by SiC-based neutron power monitors that will include but not be limited to prospects for on-line fault identification and diagnosis using pulse height and pulse shape analysis and the use of miniature SiC detectors to define axial, azimuthal, and radial flux profiles.

(3) A prototype SiC-based, neutron-power monitor with high event rate electronics whose performance will be evaluated in the Ohio State University Research Reactor under neutron fluence rate conditions that provide pulse rates that are commensurate with monitoring requirements in both the IRIS and the GT-MHR.

NUCLEAR ENERGY RESEARCH INITIATIVE

## 5. Advanced Nuclear Fuels/Fuel Cycles

This element of the program includes 16 NERI research projects to date of which 8 were awarded in FY 1999, 1 in FY 2000, 1 in FY 2001, and 6 in FY 2002. It includes research and development to provide measurable improvements in the understanding and performance of nuclear fuel and fuel cycles with respect to safety, waste production, proliferation-resistance, and economics, in order to enhance the long-term viability of nuclear energy systems. This effort includes enhanced performance of fuels for advanced systems, and development of fuels capable of withstanding the conditions in the supercritical LWR regime and of advanced proliferation-resistant fuels capable of high burn-up such as those needed in support of the Generation IV concepts.

The scope of this long-term R&D encompasses a variety of thermal and fast spectrum power reactor fuel forms, including ceramic, metal, hybrid, (e.g., cermet, cercet), and liquid, as well as such fuel types as oxides, nitrides, carbides, and metallics. Enabling technologies such as advanced cladding, water chemistry, and alternative moderators and coolants are also considered. The fuel cycle research includes consideration of advanced enrichment technologies for fuel and burnable absorbers and considers the impact of fuel cycle options on the proliferation of nuclear weapons materials. R&D topics also include development of higher density LEU (<20 percent U-235) fuels for research and development reactors.

Currently selected projects include innovative concepts for the following:

- Material preparation and production of nuclear fuels
- Inherently safe fuel designs and core response
- Study of life-limiting phenomena for high burn-up or long life fuels
- High temperature fuel and material performance
- Critical safety data and reactor physics data for advanced fuel compositions and enrichments above five percent
- Innovation in fuel design, composition, or other attributes that maximize energy production, optimize fissile material utilization, or reduce production costs

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Advanced Nuclear Fuels/Fuel Cycles Project Summaries and Abstracts

NUCLEAR ENERGY RESEARCH INITIATIVE

# Development of Improved Burnable Poisons for Commercial Nuclear Power Reactors

Primary Investigator: M.L. Grossbeck, Oak Ridge National Laboratory

Project Number: 99-074

Project Start Date: August 1999

Project End Date: June 2003

## Research Objectives

Burnable poisons (BPs) are materials that are strong absorbers of neutrons but transmute to weak absorbers upon absorption of a neutron. Such materials are used in nearly all nuclear reactors to level the power distribution and to aid in reactivity control. Burnable poisons will become even more important in Generation IV reactors where a blend of various materials with differing absorption cross sections and, therefore, lifetimes, will be necessary to permit high initial fuel loading, making long-life cores possible.

Burnable poisons used at the present time suffer from two disadvantages. The first is that boron, which is widely used, transmutes to helium, which creates undesirable internal fuel pin pressures. The second is that all other materials do not burn, or transmute, fully, and thus some absorbing material remains at the end of fuel life. This limits the amount of fuel that can be used, resulting in less efficient operation than could otherwise be achieved. Elimination or reduction of these two effects will lead to higher fuel burnup, resulting in lower cost of operation.

For many absorbing elements, such as gadolinium, it is isotopes other than the primary absorber that lead to the undesirable residual reactivity. The objective of this research is to identify single isotopes that can be used as burnable poisons that would not remain in significant quantities at the end of core life. State-of-the-art computer codes are being used to model a pressurized water reactor core with various burnable poison configurations. The second phase of the project is to separate isotopes of candidate elements to determine the enrichment attainable, the annual production, and the cost. The third phase of the project investigates compatibility of the absorber materials with the fuel and identifies potential difficulties in their use.

## Research Progress

The progress of each phase of the project will be discussed in turn.

Identification of Candidate Isotopes:  The first phase of the project is complete in that isotopes have been identified as potential advanced burnable poisons. The isotopes $^{157}Gd$, $^{149}Sm$, $^{167}Er$, $^{164}Dy$, $^{177}Hf$, and $^{151}Eu$ have all been identified and studied as candidate burnable poisons.

For this analysis, a 3,400-MWth pressurized water reactor (PWR) with a 17 x 17 array of fuel rods per assembly was modeled. Cases of BP loading in 8, 16, 64, and 104 fuel rods were studied in four configurations:

- BP homogeneously mixed with fuel
- BP mixed in the outer one third of the fuel pellets
- A thin coating of BP on the outside surface of the fuel pellets
- BP metal alloyed with the cladding

A fuel enrichment of 4.5 percent was used for most cases in an effort to achieve a four-year fuel cycle, although an enrichment of 6 percent was studied in a few cases to increase the length of the fuel cycle.

Three-dimensional neutronics calculations were performed using a sequence of the MCNP4C, Tally, and Origen2 codes in order to generate fine group fluxes and cross sections. The resulting zone-dependent fluxes and cross sections were then used to generate power and burn-up distributions.

A fuel cycle length of four years was selected for study with the residual negative reactivity at the end of the cycle of primary interest. This residual absorber penalty (RAP), expressed in terms of days of operation, was calculated for each isotope and configuration of interest. By comparing the RAP for naturally occurring elements and separated isotopes, promising isotopes were identified. Figure 1 shows a plot of negative reactivity due to the BP at the beginning of life (BOL) as a function of the RAP for the case of Gd in 16 fuel rods.

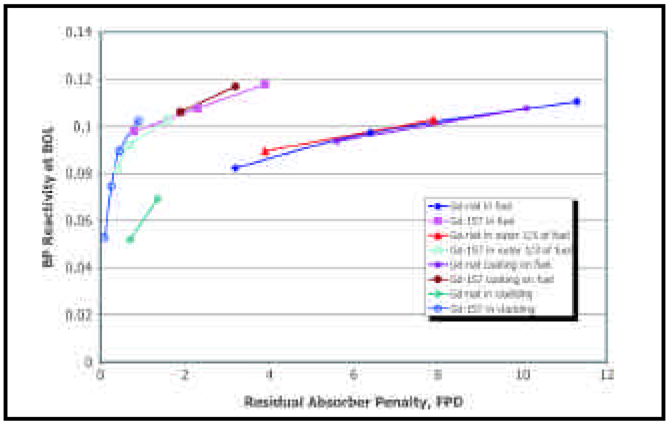



Figure 1. The graph illustrates negative reactivity from burnable poison at beginning of life as a function of the residual absorber penalty in full power days. The case is for Gd in 16 fuel rods in four configurations.

For an initial negative reactivity of -0.1 $\delta k/k_{eff}$, the difference between the two families of curves indicates a savings of six days of operation by the use of Gd-157 instead of natural Gd. This would achieve a savings of approximately $6 million. Similar analyses indicate that Sm-149 results in a savings of 44 days and Er-167 in a savings of 36 days.

An interesting result seen from Figure 1 is that the RAP for a given initial negative reactivity is independent of the configuration for a given number of fuel rods. The initial reactivity is, of course, strongly dependent upon the configuration due to self-shielding effects.

The RAP is not the only parameter of interest. Selection of a burnable poison depends upon such properties as the burn-out rate and the initial negative reactivity. A BP that burns out within the first month would be nearly useless, and a very high negative reactivity could result in a positive void coefficient. The burn-out rate is illustrated in Figure 2 in terms of the reactivity remaining at 120 days, one year, and four years. It can be seen that natural Gd is satisfactory, although improved by isotope separation. Sm becomes promising when only Sm-149 is present, and Er-167 also becomes a candidate. Although not shown in Figure 2, Er-167 has improved burnout time dependence and a lower RAP than Gd if it is mixed into only the outer one-third of the fuel.

<u>Separation of Isotopes:</u> The plasma separation process is to be used to demonstrate the separation of candidate

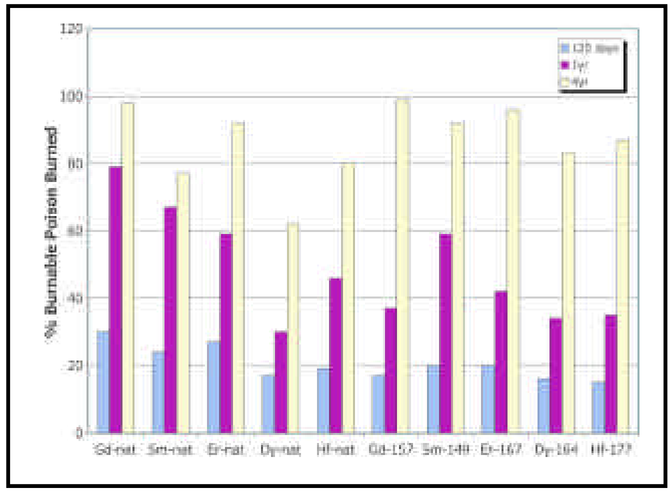



Figure 2. The time dependence of burnable poison depletion is shown, comparing naturally occurring elements and separated isotopes for the case of the BP mixed in the fuel as an oxide in eight fuel rods.

isotopes. This phase of the project was delayed because of unavailability of the separation plant, which is only now in the shake-down phase. However, fabrication of a gadolinium target for the plasma process is nearly complete. Ingots of approximately 20 kg each of Gd and Dy have been melted and cast into a large ingot. The ingot was sliced into 3.2 mm slabs by electro-discharge machining. The slabs were then welded into a single plate 0.61 x 0.58 m. This plate is now being plasma sprayed with copper on one side after which cooling coils will be soldered onto the copper-coated surface. This plate will then serve as the source target for the plasma separation process.

<u>Compatibility of Absorber Materials:</u> Considerable work has already been done by other investigators on the compatibility of rare earth oxides, especially gadolinium oxide, with uranium oxide. This work has identified reduction in thermal conductivity as the primary disadvantage in incorporating burnable poisons with fuel. Separation of isotopes permits lower concentrations of burnable poisons to be used, and thus tends to ameliorate the problem.

As was previously mentioned, this study suggests incorporating burnable poison in cladding as one option. Making a homogeneous alloy is generally easier than blending a ceramic, and incorporating the burnable poison in the fuel cladding, where the thermal flux is higher than in the fuel, permits faster and more complete burn-up. A patent disclosure for this invention has been filed (DOE Docket S-99,217; UT Battelle Docket ID 1044). Several scoping alloys of zirconium and Gd, Dy, Sm, and Er have been prepared. Bend tests have indicated no serious embrittlement resulting from the introduction of the rare earths. Larger heats of Zircaloy-rare earth alloys have been prepared in anticipation of corrosion testing.

## Planned Activities

The actual separation of isotopes is a major part of the project that has been seriously delayed because of unavailability of the plasma separation plant. This was a known uncertainty from the beginning, but the plant is now partially operational. Because of this delay, a no-cost extension of the project through June 2003 has been approved by the DOE. Preparations of a target proved to be far more difficult than anticipated, but a gadolinium target is now nearing completion. Methods developed for gadolinium are expected to apply to other rare earths, although time and funds may preclude all but perhaps one additional element. A separation run is expected to be made in the next two months.

The Zircaloy-rare earth alloys will be fabricated into the proper specimens for corrosion testing. Preparations are being made for high -pressure water and steam corrosion testing.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Fuel for a Once-Through Cycle - $(Th,U)O_2$ in a Metal Matrix

**Primary Investigator:** Sean M. McDeavitt, Argonne National Laboratory

**Collaborators:** Purdue University

**Project Number:** 99-095

**Project Start Date:** August 1999

**Project End Date:** December 2002

## Research Objectives

This project seeks to combine the advantages to be gained from metal-matrix cermet nuclear fuel with the resource-extension potential of the thorium oxide fuel cycle and the inherent proliferation resistance of mixed oxide ceramics. The approach involves fuel pins containing $(Th,U)O_2$ microspheres dispersed in a zirconium metal matrix that can achieve high burn-up and be directly disposed in a once-through fuel cycle. The beneficial aspects of the high conductivity fuel may also enable fuel assembly and reactor designs that support advanced boiling or supercritical water concepts. These advantages fit well with the Department of Energy's focus on the development of Generation IV nuclear power systems and proliferation-resistant fuel cycles.

## Research Progress

Figure 1 shows a diagram of the proposed cermet nuclear fuel concept. Cermet fuels have demonstrated the ability to enhance fuel performance and reactor safety because their high-conductivity matrix maintains low internal temperatures, which restrains fuel performance limiting phenomena and minimizes stored energy in the fuel pins. The combination of these benefits with the inherent proliferation resistance, high burn-up capability, and favorable neutronic properties of the thorium fuel cycle produces intriguing options for advanced nuclear fuel cycles. The fuel "meat" is composed of a fine dispersion of $(Th,U)O_2$ microspheres that have a theoretical density between 70 percent and 99 percent and a uranium enrichment below 20 percent U-235. Nominal values for the microsphere diameter, $ThO_2$-to-$UO_2$ ratio, fuel-to-matrix ratio, and U-235 enrichment were selected as approximately 50 μm, 50:50, 50:50, and approximately 19.5 percent, respectively, to provide guidance for the calculation and experimental activities carried out within the project. Important project achievements include (1) simulations of the core design and fuel cycle, (2) creation of a detailed thermal model for cermet fuels, (3) establishment of laboratory-scale fabrication equipment for $(Th,U)O_2$ microspheres by spray drying and sintering, and (4) establishment of laboratory-scale fabrication equipment for the powder-in-tube drawing of cermet rods.

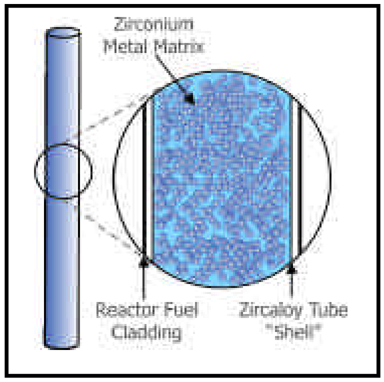



Figure 1. The schematic is a conceptual sketch for the $(Th,U)O_2$ Dispersion Fuel Pin.

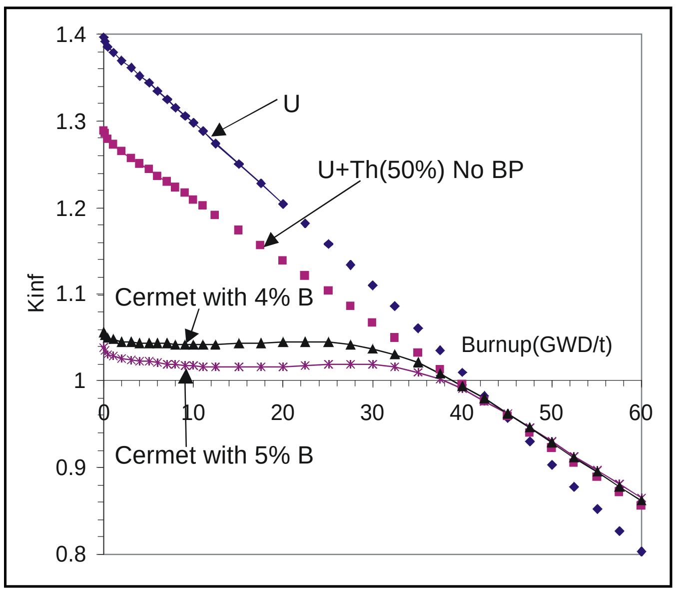



Figure 2. The graph illustrates inverse multiplication (1/M) plot for the first Burn-up Credit critical assembly. The final fuel rod configuration for this core is shown in the photographic insert.

Neutronics calculations were performed using the commercial lattice physics code HELIOS from Studsvik/Scandpower. Benchmarking calculations with Monte Carlo burn-up codes were used to demonstrate that HELIOS was effective for modeling thorium fuel lattices. The initial baseline simulations of the $(Th,U)O_2$ cermet used variations of the nominal fuel description (above) to generate comparisons with typical reactor fuels and to perform parametric studies. An increased-diameter ("fat") fuel pin in a tight lattice boiling water reactor (BWR) had a discharge burnup of approximately 80 MWd/kgHM using a U-235 enrichment of about 10 percent in a pincell calculation. A tight-pitch hexagonal lattice was then benchmarked and studied for a BWR core. Because of the substantial boiling in a BWR core, there is a natural hardening of the neutron spectrum with a corresponding increase in the fuel conversion ratio. Using the nominal (Th,U)O2 cermet, the moderator-to-fuel area was reduced in the simulated core and the conversion ratio increased from approximately 0.6 to more than 0.9, which resulted in a substantial increase in the fuel burnup over the initial result.

One notable calculation that was made is illustrated in Figure 2. In this simulation using a standard 8x8 BWR lattice, burnable absorbers are designed into the fuel matrix (as either B4C or ZrB2) with a cermet composition of 40 vol. percent Zr and 60 vol. percent heavy metal. Two types of heavy metal loadings were examined, one with only uranium and the other with mixed 50 percent thorium and 50 percent uranium (10 percent U-235 enrichment). The net fissile pin enrichment was approximately 5 percent in both cases. Two results are shown with four of the special burnable absorber rods present in the 8x8 assembly; these rods were doped with boron in the zirconium metal matrix at concentrations of 4 and 5 atom percent. The use of 4 rods at 5 percent boron very nearly controls all the excess reactivity in the assembly with no significant penalty to the achievable burn-up. This is an important result which may enable autonomous control in future reactor designs.

The high thermal conductivity of the zirconium matrix greatly enhances heat removal; thus, the centerline fuel temperature will be significantly lower than that of a monolithic ceramic fuel pin. This point is important because the lower overall fuel temperature reduces the performance-limiting impact of fission product migration, fuel swelling, and other in-reactor phenomena. In addition, the high-conductivity matrix results in a low stored energy content due to the low internal fuel temperatures, which contributes to severe accident mitigation and a low fuel failure rate. A detailed thermal model has been developed based on the effective conductivity across an ideal interface to simulate the behavior of cermet fuel.

Spray drying is a physical process for granulating fine powders that is widely used in the chemical, pharmaceutical, ceramic, and food industries. The spray drying system (Figure 3a) is a commercial laboratory-scale spray dryer made by Niro Inc., and it consists of a funnel-shaped chamber approximately 1m in diameter with an insulated stainless steel double wall. Parametric studies were carried out to formulate stable, dense, and homogeneous aqueous slurries of urania and thoria powders for the production of microspheres. The parameters studied include (a) particle size distribution after ball-milling; (b) slurry viscosity; (c) zeta potential; (d) slurry flowability, stability and cleanability; (e) microsphere green strength; and (f) effects of organic dispersants. In an initial spray drying run, a slurry of $(U,Th)O_2$ was prepared by wet-milling fine urania and thoria powders and 0.5 volume percent triethanolamine for 24 hours (hydrochloric acid was used to adjust the pH of the slurry to 3 and the volume of the slurry was about 50 ml). A representative picture of the final sintered microspheres is shown in Figure 3b.

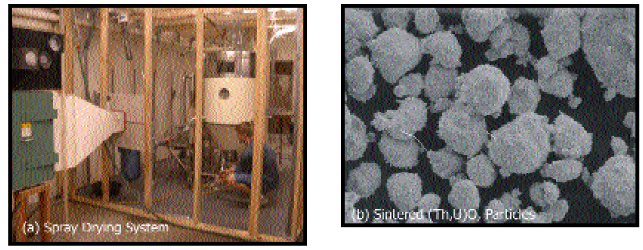



Figure 3. Components of the Spray Drying Process are pictured: (a) Photograph of the spray drying system in its enclosure with a HEPA filtration system, and (b) Electron micrograph of spray-dried $(U,Th)O_2$ microspheres sintered at 1,650°C for 10h in flowing Ar-5 percent $H_2$.

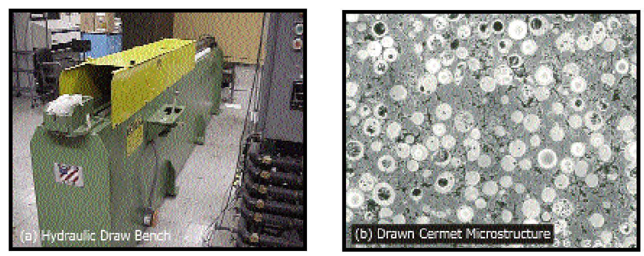



Figure 4. Components of the Powder-in-Tube Fabrication Process are shown: (a) Photograph of the hydraulic draw bench and (b) Electron micrograph of tungsten carbide microspheres in a zirconium matrix fabricated by powder-in tube drawing.

The powder-in-tube fabrication method is being developed as a simple low-temperature alternative to elevated temperature methods. In this process, oxide microspheres (50 to 1,000 μm diameter) and zirconium metal powders (~44 μm nominal diameter) are dry-mixed and loaded into stainless steel or Zircaloy drawing tubes and vibratory-packed. The powder-containing tube is drawn through a die to reduce the diameter and compact the powder into a dense matrix. The metals are annealed between 500°C and 1,000°C to remove strain hardening and strengthen interfacial bonding. Multiple cycles are used with sequentially decreasing die sizes to achieve complete densification. A hydraulic draw bench, manufactured by Fenn, has been installed in a radioactive materials lab (Figure 4a) alongside a materials research furnace. Initial fabrication demonstrations were completed using non-radioactive surrogate materials and a representative cermet cross-section is shown in Figure 4b.

## Planned Activities

This project is nearly complete. Remaining efforts will include completing the neutronic modeling activities and fabricating a representative cermet fuel pin using spray dried $(Th,U)O_2$ microspheres. The modeling work is focussed on applying the unique advantages of this novel fuel form toward high-conversion reactor concepts including BWR, pressurized water reactor, reduced moderator water reactor, and supercritical light water reactor systems. The $(Th,U)O_2$ microspheres have been manufactured and the final product is scheduled to be drawn in November 2002.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Fundamental Mechanisms of Corrosion of Advanced Light Water Reactor Fuel Cladding Alloys at High Burn-Up

Primary Investigator: Randy G. Lott, Westinghouse Electric Company LLC

Collaborators: Pennsylvania State University; Argonne National Laboratory (West); Idaho National Engineering and Environmental Laboratory

Project Number: 99-128

Project Start Date: August 1999

Project End Date: January 2003

## Research Objectives

The corrosion behavior of nuclear fuel cladding is a key factor limiting the performance of nuclear fuel elements. Improved cladding alloys, which resist corrosion and radiation damage, will facilitate higher burn-up core designs. The objective of this study is to understand the mechanisms by which alloy composition, heat treatment, and microstructure affect corrosion rate. This knowledge will be used to predict the behavior of existing alloys outside the current experience base (for example at high burn-up) and predict the effects of changes in operational conditions on zirconium alloy behavior.

Zirconium alloys corrode by the formation of a highly adherent protective oxide layer. The working hypothesis of this project is that alloy composition, microstructure, and heat treatment affect corrosion rates through their effect on the protective oxide structure and ion transport properties. Therefore, particular emphasis has been placed on detailed characterizations of the oxides formed on a series of experimental alloys. The experimental task in this project is to identify these differences and understand how they affect corrosion behavior. To do this, several microstructural examination techniques are being employed, including transmission electron microscopy (TEM), electrochemical impedance spectroscopy (EIS), and a selection of fluorescence and diffraction techniques using synchrotron radiation at the Advanced Photon Source (APS).

Detailed characterizations of oxides are only useful if the observations can be linked to the corrosion behavior of the alloy. That link requires a model of the corrosion mechanism. The modeling effort is designed to organize the data from the characterization studies in a self-consistent manner and link those observations to the corrosion behavior. The ultimate objective of this study is a linkage between the characterization and theoretical modeling efforts that will produce improved alloy specifications.

## Research Progress

Collaboration among the various members of the research team has led to the development of innovative descriptions of the corrosion process. The most promising of these ideas have been built into the corrosion model. A relevant set of alloys has been selected that bracket the expected range of corrosion behavior and that will be used both for the experimental characterization and for the modeling effort. The testing has been expanded to include oxide produced in steam and Li environments.

A systematic series of experiments have been performed at APS and the TEM characterization of oxides is continuing. Both microfluorescence compositional information and diffraction have been obtained using a unique x-ray microprobe facility, as shown schematically in Figure 1.

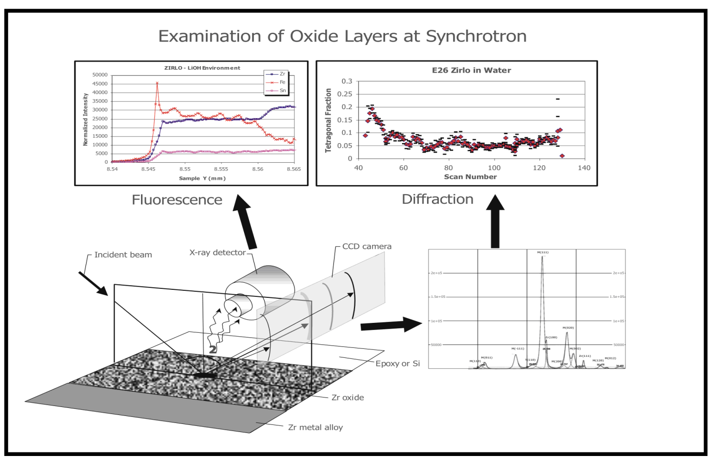



Figure 1. Oxide layers are examined in cross section at APS, and both microfluorescence and microdiffraction information can be obtained, with a resolution below 0.3 micron. Examples of real data obtained using this technique are shown in the graphs above.

Various oxides formed in various alloys and under three different environments have been characterized with respect to their tetragonal-to-monoclinic ratio and alloying element distribution in the oxide layer using synchrotron radiation. Cross-sectional TEM samples have been examined and more detailed work will be performed that will serve as complementary information to the synchrotron work.

The initial certification and experiments with EIS have been completed and data has been collected. The EIS results are consistent with the existence of a porous outer oxide and a dense, insulating inner oxide. The results have been analyzed in an attempt to understand the electronic differences in the two layers. The noted variations did not appear to be significant. Newly developed optical techniques have proven to be more efficient at identifying the layered structures of the oxides. Therefore, the EIS studies have been scaled back in favor of more detailed TEM and APS evaluations.

The autoclave design for the electrochemistry and radiolysis studies was completed. However, in accordance with the modified program plan, no further development of this facility will be undertaken in Year 3 of the Program.

The model engine, which connects the various model components and tracks the evolution of both the continuous and porous oxides, has been developed. This engine also includes a hydrogen accumulation mechanism. The engine has been developed as a Visual Basic Application, which writes the output directly into an Excel spreadsheet.

A schematic of the modeling process is indicated in Figure 2. Oxide growth is simulated using suitably small time steps. The environmental factors controlling the growth rate and critical thickness are calculated at each increment. These factors are then used as inputs to the growth rate and critical thickness modules. The

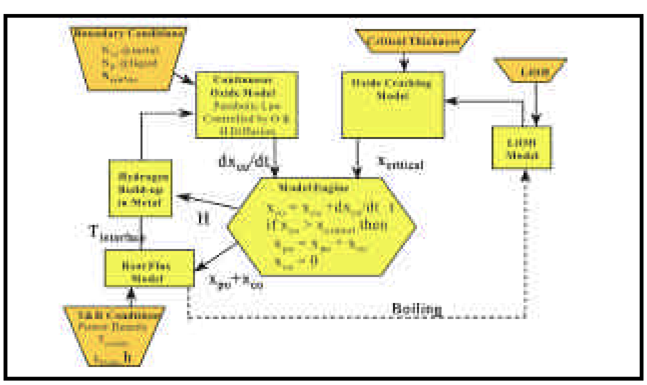



Figure 2. The schematic shows the application of the model to calculating oxide thickness.

continuous oxide growth rate and the critical thickness are fed into the model engine. The basic model engine is used to determine the oxide thickness as a function of time. The engine incrementally increases the continuous oxide thickness and then checks to see if the thickness exceeds the critical value. When the continuous oxide measurement exceeds the critical value, the entire thickness is added to the porous oxide thickness and the continuous oxide thickness is returned to zero. The model is structured to assure consistency in the inputs to all modules. A single set of external inputs, describing the alloy and the reactor operating conditions, is used for all modules.

The primary task in designing an advanced alloy is understanding how each of the potential alloy constituents affect the corrosion behavior. Both the composition and the heat treatment of the material determine the corrosion behavior of the alloy. Therefore, it is important to understand how the microstructure of the alloy controls oxide formation. Factors controlling corrosion have been identified as the fraction of the alloying element precipitated, and the precipitate size. Lessons learned from the corrosion model have provided insight into these processes. As more is learned about the behavior of each alloying element, it should become possible to use the model as a tool for designing new alloys.

## Planned Activities

During the final quarter of Budget Year 2, preparatory work has been undertaken for several new initiatives. A change in the workscope was negotiated to allow TEM examination of the irradiated specimens at Argonne National Laboratory. The APS runs have produced a large volume of diffraction data that is still being analyzed. Revisions to the model that will more accurately describe the oxidation of second phase particles within the growing oxide film are being developed.

In the final Budget Year of the project, it is expected that the combined knowledge derived from the various parts of the project will allow the design of new alloys for extended burn-up applications. Basing the design of the new alloys on mechanistic knowledge will be a big step forward to ensure reliable fuel operation at high burn-up rates.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Advanced Proliferation-Resistant, Lower-Cost, Uranium-Thorium Dioxide Fuels for Light Water Reactors

Primary Investigator: Philip E. MacDonald, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Argonne National Laboratory; University of Florida; Framatome ANP; Korea Atomic Energy Research Institute; Massachusetts Institute of Technology (MIT); Purdue University; Westinghouse Electric Corporation

Project Number: 99-153

Project Start Date: August 1999

Project End Date: August 2003

## Research Objectives

The overall objective of this NERI project is to evaluate the efficacy of high burn-up mixed thorium-uranium dioxide ($ThO_2$-$UO_2$) fuels for light water reactors (LWRs). A mixed thoria-urania fuel that can be operated to a relatively high burn-up level in current and future LWRs may have the potential to improve fuel cycle economics (allow higher sustainable plant capacity factors), improve fuel performance, increase proliferation resistance, and be a more stable and insoluble waste product than $UO_2$.

The project has been organized into four tasks:

(1) Task 1 will consist of fuel cycle neutonics and economics analysis to determine the economic viability of a $ThO_2$-$UO_2$ fuel cycle in PWRs.

(2) Task 2 will determine whether or not $ThO_2$-$UO_2$ fuel can be manufactured economically.

(3) Task 3 will evaluate the behavior of $ThO_2$-$UO_2$ fuel during normal, off-normal, and accident conditions and compare the results with the results of previous $UO_2$ fuel evaluations and U.S. Nuclear Regulatory Commission (NRC) licensing standards.

(4) Task 4 will determine the long-term stability of $ThO_2$-$UO_2$ waste.

## Research Progress

Progress made on each of the tasks will be described in turn in this section.

Task 1 - Reactor Core Analysis and Fuel Cycle Design: Due to the relatively poor economic performance of the homogeneously mixed urania-thoria fuel, the focus of the physics work under Task 1 during Year 2 and the first part of Year 3 was primarily on the performance and economics of using micro-heterogeneous fuel forms, where some small distance physically separates the uranium and thorium. When compared to the equivalent homogeneous case (i.e., the same urania-thoria weight percentages), an increase in burn-up is observed, which improves the economics of using thorium-based fuels. However, the economic improvement due to the use of any of the various micro-heterogeneous fuel forms is not sufficient to compensate for the costs of the increased Separative Work Units (SWUs) required for thorium oxide fuels. Therefore, the work at Framatome, INEEL, and MIT during most of Year 3 has focused on use of thorium to burn unwanted reactor or weapons grade plutonium.

Framatome ANP has completed the design of a weapons grade plutonium-thorium oxide-fueled core. Three different fuel pin loadings of plutonium are used to minimize the power peaking in the corner pins and in the pins near the water holes. The design meets current thermal-hydraulic and safety criteria. A lifetime depletion calculation was completed and the plutonium-thorium fuel design was found to be as reactive as plutonium-uranium cores. However, in the case of the plutonium-thorium core, less than 10 percent of the original plutonium inventory remains at 50,000 MWd/MtHM. The analysis of selected reactor transients demonstrated that it is feasible to license and operate safely a reactor fueled with plutonium-thorium blended fuel. In most cases analyzed, the thorium mixture had less severe consequences than those for a core comprised of Low Enriched Uranium (LEU) fuel. In the analyzed cases where the consequences were more severe, they were still within acceptable limits.

The INEEL has completed analyses of a plutonium-thorium oxide-fueled core with every other fuel rod LEU $UO_2$ and plutonium-thorium oxide. These preliminary analyses have shown that the combination of thorium, recovered uranium, and reactor grade plutonium in an oxide fuel for light water reactors can be an effective way to consume reactor grade plutonium and to transmute the remaining plutonium isotopes into a mixture that is more proliferation-resistant, while still allowing the reactor reactivity control systems to remain similar to that for $UO_2$ cores.

MIT researchers benchmarked the analysis tools CASMO4 and MCODE against the IAEA standard problem for analysis of plutonia-thoria lattices. They then used those codes to perform a comprehensive study of the reactor grade plutonium destruction capabilities of homogeneously mixed $PuO_2$-$ThO_2$ fuels in LWRs as a function of H/HM ratios. For the un-denatured cases, up to 1,000 kg of plutonium can potentially be destroyed per GWe-yr. The residual plutonium fraction (relative to the initially loaded Pu) in discharged fuel can be minimized by increasing the H/HM ratio and can potentially be as low as 25 percent. However, denaturing of mixed plutonium-thorium oxide fuel impairs the plutonium destruction effectiveness by about 20 percent. This penalty can also be minimized by increasing the H/HM ratio. The results of the reactivity coefficient evaluations indicate that mixed plutonium-thorium oxide fuel can be used for plutonium disposition in conventional PWRs with some changes in reactor reactivity control systems. The Doppler coefficient, boron worth, and $\beta_{eff}$ of the plutonium-thorium oxide fuel are of about the same order as plutonium-uranium oxide fuel values.

<u>Task 2 - Fuel Manufacturing Costs:</u> This task was organized into three major activities: an engineering study of the feasibility of producing the thorium-uranium fuel in current nuclear fuel production facilities, an effort to estimate the cost of fabricating $ThO_2$-$UO_2$ oxide fuel, and a developmental effort to make fuel pellets with appropriate densities and to use this material to determine fundamental heat transfer properties to use in the modeling efforts. The Westinghouse Electric Company completed the first two tasks at the end of Year 2 and the results were reported in the 2$^{nd}$ Annual Progress Report. Purdue University, with help from Westinghouse, is continuing to evaluate the fabrication issues associated with co-precipitation of the powder and with pressing, sintering, and grinding $ThO_2$-$UO_2$ fuel pellets. Purdue University is also tasked with making various material measurements to support the modeling efforts.

<u>Task 3 - Fuel Performance:</u> This task is providing tools to evaluate the thermal, mechanical, and chemical aspects of the behavior of $ThO_2$-$UO_2$ fuel rods during normal, off-normal, and design basis accident conditions. During Year 3, MIT has been developing a version of the transient code FRAP-T6 for analyses of $ThO_2$-$UO_2$ fuel behavior during a Reactivity Initiated Accident (RIA) event. Modifications to FRAP-T6 included the addition of thoria fuel properties (heat capacity, thermal expansion, thermal conductivity); a low temperature burst stress model; and the gaseous swelling contribution to the cladding strain. The available high burnup $UO_2$ fuel tests have been analyzed and the modified FRAP-T6 seems to reasonably predict the residual cladding strains in the tests. Analyses of $ThO_2$-$UO_2$ fuel in a typical LWR indicate that it will tend to perform better than $UO_2$ fuel under RIA event conditions due to its lower thermal expansion and flatter power distribution in the fuel pellet (less power and less fission gas in the rim region).

During Year 2, MIT researchers calculated that the most promising micro-hetrogeneous thoria-urania arrangement with respect to achievable burnup is the axial micro-heterogeneous design with $UO_2$ and thorium section lengths of 4 and 8 cm, respectively. This design increases the fuel discharge burnup by a significant amount over the $UO_2$ base case, about 13 percent to 15 percent. In addition, this design offers the benefit of a substantial reduction of poison to compensate for the reactivity excess at beginning-of-life. Although this design manifests appreciable neutronic advantages, the absence of fissile material in the $ThO_2$ section at beginning-of-life results in large local power peaking. The most effective way to reduce the local peaking is to add uranium with fissile U-235 into the $ThO_2$ section. However, because homogeneous mixing of uranium in the thorium slug significantly impairs the reactivity-limited burnup performance, a modified axial and radial micro-heterogeneous fuel pin design (DuUAx4) was developed by introducing a 25 vol percent central void in the $UO_2$ driver zone and moving the extra $UO_2$ into the blanket zone as an inner ring with $ThO_2$ as an outer ring.

During Year 3, calculations were performed at the INEEL to compare the temperature behavior of DuUAx4 and conventional 100 percent $UO_2$ fuel rods during a large break LOCA. The calculations were performed with the SCDAP/RELAP MOD3.3 code extended for the analysis of $ThO_2$-$UO_2$ fuel rods and extended for the modeling of axial heat conduction. A solution scheme for temperature involving either a fixed or moving fine mesh was added to the SCDAP/RELAP5 computer code. The fine mesh allows for mesh sizes in the axial direction as small as 1 mm (order of the thickness of the cladding). This small mesh size provides for an accurate calculation of cladding temperatures in the vicinity of the quench front or at the axial interface of a seed and blanket region in a fuel rod. It was found that the maximum cladding temperature of the DuUAx4 fuel during a LOCA is not significantly greater than that in conventional 100 percent $UO_2$ fuel.

Task 4 - Long Term Stability of $ThO_2$-$UO_2$ Waste: This research is focused on measuring uranium dissolution from $(U,Th)O_2$ solid solutions as a function of the uranium content to determine the degree to which the mixed oxide is superior to $UO_2$ as a waste form. Dissolution studies on irradiated and unirradiated $(U,Th)O_2$ pellets and pellet fragments are underway. Irradiated fuels under investigation range in composition from 2 to 5.2 percent $UO_2$. This work is being conducted at Argonne National Laboratory East (ANL E). The experiments on the unirradiated fuels at the University of Florida involve compositions of from 5 to 50 percent $UO_2$. Dissolution behavior is being studied in J13 well water at both 90ºC and room temperature. The results of this study will include (1) comparison of the dissolution behavior of irradiated and unirradiated fuel (nominal composition of 5 percent $UO_2$) to determine the effect of fuel burnup on dissolution and (2) the effect of solid solution composition on the dissolution behavior of the unirradiated fuel. Studies of the dry oxidation behavior of $(U,Th)O_2$ are also continuing. The experiments involve gravimetric analysis of sample powders heated in an oxidizing or reducing environment. The results will show the oxidation behavior of $(U,Th)O_2$ as a function of the solid composition, and give an indication of the effect thorium has on the oxidation of uranium.

Task 5 - Korean Work: The Koreans have been working on four tasks: core design analyses, fuel pellet manufacturing technologies, fuel rod performance analysis, and xenon diffusivity measurements. In the area of core design analyses, the Koreans completed their analysis of the mixed core concept of duplex $(Th,U)O_2$ and $UO_2$ fuels. The uranium ore and SWU costs per MWD for the mixed core of thorium-uranium oxide with uranium oxide fuel are improved over the costs for a homogeneous thoria-urania core. However, even with a long fuel cycle scheme, the thorium-based mixed core concept does not show a superior potential in fuel economics to an all-uranium oxide core. To further improve their fuel pellet manufacturing technologies, three kinds of pellets ($ThO_2$, $ThO_2$-35%$UO_2$, $ThO_2$-65%$UO_2$) have been fabricated by powder processing. However, good $(Th,U)O_2$ pellet homogeneity could not be achieved using dry milling and a wet mill process was developed. The resulting $(Th,U)O_2$ pellets had a density greater than 95 percent TD and good homogeneity.

In the area of fuel rod performance, the INFRA-Th computer code was developed by adding the thermal conductivity, radial power, and burn-up distribution, and thermal expansion models for $ThO_2$-$UO_2$ fuel to the $UO_2$ performance analysis code, INFRA. Also, a $ThO_2$-$UO_2$ irradiation test, called IFA-652.1, was started in June 2000 in the Halden Reactor. The test rod is instrumented with a thermo-couple and pressure transducer. The fuel centerline temperature and rod internal pressure data from IFA-652.1 has been compared with predictions from the INFRA-Th computer code. Four xenon diffusivity measurement experiments have been completed during Year 3 using natural UO2. The diffusion coefficients measured in this work for xenon in $UO_2$ are consistent with the published literature. Work with $ThO_2$-$UO_2$ fuel will start shortly.

## Planned Activities

All of the planned work is complete in Tasks 1, 2, and 3 of this NERI project except for the thoria-urania fuel property measurements that were to have been done by Purdue University. It should be noted that significantly more work was done in Task 1 by Framatome ANP and MIT than was originally proposed and planned. The waste studies in Task 4 at the University of Florida and ANL are on schedule for completion by the end of Year 3. The work in Korea is not directly funded by the Department of Energy; however, all the work originally agreed to has been carried out as planned and is essentially complete.

NUCLEAR ENERGY RESEARCH INITIATIVE

# A Proliferation-Resistant Hexagonal Tight Lattice BWR Fuel Core Design for Increased Burn-up and Reduced Fuel Storage Requirements

Primary Investigator: Hiroshi Takahashi, Brookhaven National Laboratory

Collaborators: Purdue University; Korean Atomic Energy Research Institute (KAERI); University of Mining & Metallurgy (Poland)

Project Number: 99-164

Project Start Date: August 1999

Project End Date: September 2002

## Research Objectives

The major objective of this project is to advance the well-developed, water-cooled reactor technology in order to make efficient use of the abundant thorium resource available in the earth's crust. Considerable effort has been invested in development of the sodium-cooled fast reactor to breed fissionable $^{239}$Pu from natural uranium. Much less effort has been expended into development of alternative technologies to safely and efficiently make use of the thorium resource. This project will investigate the feasibility of a plutonium-thorium (Pu-Th) fuel cycle for a new type of high conversion reactor cooled by boiling water (HCBWR). The technology will be developed to burn existing stocks of plutonium, while converting the fertile thorium to fissile $^{233}$U. The high conversion rate will take place in a fast neutron spectrum through the design feature that minimizes the volume of water in very tight fuel assembly lattices. A segmented core design will be used, consisting of multiple radial and axial zones, and the design will burn the plutonium and will convert thorium into the more proliferation-resistant $^{233}$U. High fuel burn up will be possible as a result of the continuous generation and fission of $^{233}$U as the plutonium is consumed. Inherent safety will be designed into the reactor through the use of thorium as fertile material. This will introduce a negative void coefficient for those accident sequences which result in off-normal coolant boiling.

Thus, the major technical objective of the proposed project is to develop a reactor design which will accomplish the following:

- Minimize the potential for proliferation of weapons grade fissionable materials
- Maximize the inherent safety features of the reactor
- Maximize the achievable fuel burn up and plant capacity factor
- Minimize the cost of electricity generation

The research and development program proposed here will satisfy the major objectives of the NERI program:

(1) Advance the state of nuclear technology through introduction of an alternative, proliferation resistant, new type of fast reactor

(2) Advance the state of nuclear technology through extending the well-developed boiling water technologies

(3) Maintain a nuclear science and engineering capability through use of major nuclear industry reactor design and safety analysis tools during the design and safety analysis of the project

(4) Improve the safety performance of fast reactors through use of potentially safer water, which is not susceptible to combustion in air as is sodium

(5) Improve the safety of fast reactors as a result of using thorium fertile material to provide a negative void reactivity coefficient.

## Research Progress

Research has focused on neutronics design analysis of a HCBWR with Pu-Th fuel.

One of the primary innovative design features of the core proposed here is the use of thorium as fertile material. In addition to the advantageous nonproliferation and waste characteristics of thorium fuel cycles, the use of thorium is particularly important in a tight-pitch, high-conversion lattice in order to ensure a negative void coefficient throughout the operating life of the reactor.

The principal design objective of a high-conversion light water reactor (LWR) is to substantially increase the conversion ratio (fissile atoms produced per fissile atoms consumed) of the reactor without compromising the safety performance of the plant. Since existing LWRs have a relatively low conversion ratio, they require relatively frequent refueling, which limits the economic efficiency of the plant. Also, the high volume of spent fuel can pose a burden for waste storage and the accumulation of plutonium in the uranium fuel cycle can become a materials-proliferation issue. The development of fast breeder reactors (FBRs) as an alternative technology to alleviate some of these concerns has been delayed for various reasons. An intermediate solution has been to examine tight pitch LWRs that can provide significant improvements in the fuel cycle performance of the existing LWRs by taking advantage of the increased conversion ratios from the harder neutron spectrum in the tight pitch lattice, as well as by taking advantage of the waste and nonproliferation benefits of the thorium fuel cycle.

Several HCBWR designs have been proposed by researchers in Japan and elsewhere during the past several years. One of the more promising high-conversion reactor (HCR) designs is the reduced moderation water reactor (RMWR) proposed by The Japan Atomic Energy Research Institute (JAERI). Their design was based on a uranium fuel cycle and showed significant improvements in the fuel cycle performance compared to conventional BWRs. However, one of the drawbacks of their design was the potential for a positive void coefficient. In order to ensure a negative void coefficient, the JAERI researchers designed a "flat core" and introduced void tube assemblies in order to enhance neutron leakage in the event of core voiding. The use of thorium in the Purdue/BNL HCBWR design proposed here obviates the need for void tubes and makes it possible to increase the core height and improve neutron economy without the risk of a positive void coefficient. The principal reason for the improvement in the void coefficient is that Th-232 has a smaller fast fission cross section and resonance integral than U-238. In the design proposed here, it is possible to eliminate the void tubes in the RMWR design and replace the axial blanket with active fuel to increase the core height and further improve neutron economy.

The core analyses in the work here were performed with the Purdue Fuel Management Code System which is based on the Studsvik/Scandpower lattice physics code HELIOS, and the U.S. NRC core neutronics simulator, PARCS, which is coupled to the thermal-hydraulics code RELAP5. All these codes have been well-assessed and benchmarked for analysis of LWR systems.

The HCBWR developed here is characterized by a very tight lattice with a relatively small water volume fraction in the core, which therefore operates with a fast reactor neutron spectrum, and a considerably improved neutron economy compared to the current generation of LWRs. A tight lattice BWR core has a very narrow flow channels with a hydraulic diameter less than half of the regular BWR core. The tight lattice core presented a special challenge to core cooling, because of reduced water inventory and high friction in the core. The primary safety concern when reducing the moderator to fuel ratio and when using a tightly packed lattice arrangement is to maintain adequate cooling of the core during both normal operation and accident scenarios.

In the HCBWR design, the core has been placed in a vessel with a large chimney section, and the vessel is connected to the isolation cooling system (ICS). The vessel is placed in containment with a gravity driven cooling system (GDCS) and a passive containment cooling system (PCCS) in a configuration similar to General Electric's Simplified Boiling Water Reactor (SBWR). The safety systems are similar to SBWR; ICS and PCCS are scaled with power. An internal recirculation pump was placed in the downcomer to augment the buoyancy head provided by the chimney. The buoyancy provided by the chimney alone could not generate sufficient recirculation in the vessel since the tight lattice configuration resulted in much larger friction in the core than the SBWR.

The modified RELAP5 was used to simulate and analyze two of the most limiting events for a tight pitch lattice core: the Station Blackout and the Main Steam Line Break events. The constitutive relationships for RELAP5 were compared with the correlations and the data available for narrow channels, and heat transfer package was modified for narrow channel application. The results of the analyses indicate that the HCBWR system will be safely shutdown for these transients.

## Planned Activities

Evaluation of safety analyses will be continued with newly obtained core data using the RELAP5 code.

The economic and non-proliferation studies will be continued. The potential for transmutation of minor actinides using the hard neutron energy spectrum in HCBWR will be studied, as well as the deep-underground concept, which provides a safer operation than the surface reactor.

A final evaluation report is being prepared with an overview of the HCBWR system.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Development of a Stabilized Light Water Reactor (LWR) Fuel Matrix for Extended Burn-up

Primary Investigator: Brady D. Hanson, Pacific Northwest National Laboratory (PNNL)

Collaborators: University of California, Berkeley

Project Number: 99-197

Project Start Date: August 1999

Project End Date: May 2003

## Research Objectives

The main objective of this project is to develop an advanced fuel matrix based on the currently licensed $UO_2$ structure capable of achieving extended burn-up while improving safety margins and reliability for present operations. Burn-up is currently limited by fission gas release and the associated increase in fuel rod internal pressure, fuel swelling, and cladding degradation. Once fuels exceed a threshold burn-up, a "rim" or high burn-up structure (HBS) forms. The HBS is characterized by the development of a subgrain microstructure having high porosity and low thermal conductivity. It is believed that the lower thermal conductivity results in larger temperature gradients and contributes to subsequent fission gas release. Fuel designs that decrease the centerline temperature while limiting the HBS restructuring, thereby decreasing the fission gas release, should be able to achieve higher burn-up and even allow higher operating power for increased efficiency.

Research at PNNL has demonstrated that the soluble fission products and actinides present in the matrix of irradiated (spent) fuels stabilize the fuel matrix with respect to oxidation to $U_3O_8$. The higher the soluble dopant concentration, the more resistant the fuel has been to restructuring of the matrix from the cubic phase of $UO_2$ to the orthorhombic $U_3O_8$ phase. It is hypothesized that such restructuring of the uranium planes within the matrix due to oxidation is similar to the microstructural changes that occur during HBS formation. In this project, the attempt is to utilize the changes in fuel chemistry that result from doping the fuel to design a fuel that minimizes HBS formation and is more resistant to corrosion if it is ever exposed to water or air. The use of dopants that can act as getters of free oxygen and fission products (e.g, Cs, I, Tc) to minimize fuel-side corrosion of the cladding or release from the fuel matrix is also being studied.

Other fuel designs, such as radial variations in enrichment, are being examined to further minimize HBS formation. The growth of large grains, which should reduce both HBS formation and fission gas release, using a steam oxidation process, is also being studied. A combination of experimental studies and theoretical modeling is being used to determine the optimal design.

## Research Progress

Work on Task 1, Understand and Model HBS Formation, involves increasing the understanding of HBS formation in order to better design a fuel to delay or prevent its onset. A comprehensive literature review has been performed. While it is clear that no one fully understands how the HBS forms, it is becoming apparent that the accumulation of radiation damage is the main driving force. High burn-up fuels (>60 MWd/kg) have been examined at PNNL using Atomic Force Microscopy (AFM). These examinations have shown the subgrain microstructure similar to the HBS, even in the central regions of the fuel pellet where the local burn-up is below the threshold reported for bulk formation of the HBS. The temperatures near the fuel centerline are also high enough that radiation damage is readily annealed. However, the areas adjacent to pores seem to act as pinning sites for radiation damage and result in very localized restructuring. The large grain size for the advanced fuel being designed for this project should help delay HBS formation.

Most studies of HBS have utilized Electron Microprobe Analysis (EMPA) to determine concentrations of Xe, U, or Pu as a function of radial position in the fuel. In order to more fully understand the phenomena associated with HBS formation, this project has sponsored the continuing development of the Resonance Absorption Burn-up (RABURN) model at UC-Berkeley. The objective is to develop a simplified code that can, in a computationally efficient and user friendly way, provide a good approximation for calculating the radial-dependence of the fission rate and actinide production in reactor fuels. The radial dependence of soluble dopants in the $UO_2$ matrix, local temperature, and extent of radiation damage are being calculated to provide the basis for the HBS and matrix stabilization models.

Development has continued of the one-dimensional code, RABURN. In particular, it has been possible to verify most of the fundamental assumptions in the code. These assumptions are that (1) the only relevant portions of the neutron flux incident at the fuel surface correspond to those associated with the quasi-thermal and the epithermal fluxes; (2) the ratio of the integrated thermal flux to the magnitude of the epithermal flux, assumed to be of the form $\phi_o/E$, is about 0.25 to 0.35; (3) because of the 1/E variation of the epithermal flux, only the lowest-energy strong resonances need be considered explicitly; (4) because of their large mean free paths in the fuel, a neutron incident on the surface of the fuel would interact once or not at all in passing through a dimension of the fuel diameter; and, (5) the main resonances that should be considered are those in (even, even) actinides at low energies. These assumptions have all been verified by performing pin-cell calculations with the Monte Carlo code MCNP and a well-characterized spent fuel (ATM-103). RABURN has been modified to account for temperature- and burn-up-dependent thermal conductivities to calculate radial-dependent temperature distributions. The code is able to reproduce the radial-averaged concentrations of $^{235}U$, $^{236}U$, $^{238}U$, $^{237}Np$, $^{239}Pu$, and $^{240}Pu$ to within $\pm 5$ to 10 percent. However, the code still predicts Pu concentrations in the rim region will be higher by a factor of 2.7 compared to that measured. The results have also been more insensitive to parameter changes than would have been predicted. The reason for these discrepancies appears to be that the neutron flux depression at energies in the immediate vicinity of resonances in the fuel was accounted for, but the corresponding depression in the water immediately adjacent to the fuel was not.

Task 2, Develop Matrix Stabilization Model, has focused on determining the lattice parameter of both spent fuels and doped fuels produced by this project. While the team has a general understanding of how dopants stabilize the matrix with respect to oxidation, a model is being developed that accounts for these changes as a function of lattice parameter and the ionic radii and oxidation state of the dopants. Powder X-ray Diffractometry (XRD) has been used to determine the phases and lattice parameters of the fuels tested. However, the sensitivity and precision using standard quantitative XRD were not sufficient for the modeling efforts. A whole pattern fitting/Reitveld refinement program has been procured and initial results indicate much less uncertainty in the re-analyzed data.

Task 3, Design and Test Advanced Fuel Matrix, is focused on the dopant concept. A thorough literature review was performed on the effect of dopants on the $UO_2$ matrix. Additionally, a lab-scale fuel fabrication laboratory was established at PNNL and $UO_2$ and doped-$UO_2$ pellets were produced. Initial efforts have concentrated on producing homogeneously doped pellets so the results from thermogravimetric analysis (TGA) studies to examine oxidation response will be representative.

$UO_2$ powder has been procured from Framatome ANP. The $UO_2$ powder and the desired dopant are wet-milled for 24 hours to assure intimate mixing. Excess water is then removed by vacuum drying. Pellets are formed by first granulating and then pressing the mixed powder. The pellets are sintered in a 3 percent $H_2$ atmosphere at 1,650°C for 24 hours. The pellets are weighed and measured using laser dimensional analysis, and the density is determined using gas pychnometry. The pellet densities have typically been approximately equal to 95 percent of the theoretical density. The fuel pellet is then subsectioned and examined using XRD to determine lattice parameter and to verify that a solid solution was obtained. Other sections of the pellet are examined using Scanning Electron Microscopy (SEM) with an EDAX Light Element Detector and backscattered imaging to look for areas of heterogeneous dopant/fuel mixtures. It appears that homogeneous solid solution pellets can be made successfully with dopant concentrations up to at least 10 wt percent. Examples of the XRD fits to pellets with 8 wt percent $Gd_2O_3$ and 10 weigth (wt) percent $ZrO_2$, respectively, are shown in Figure 1.

## Planned Activities

The following activities are scheduled for the final year of this project:

(1) RABURN will be modified to a true cylindrical geometry and any necessary approximations to the neutron flux depression in the water will be incorporated. RABURN will then be run to determine local dopant concentrations and temperatures for comparison with AFM data to

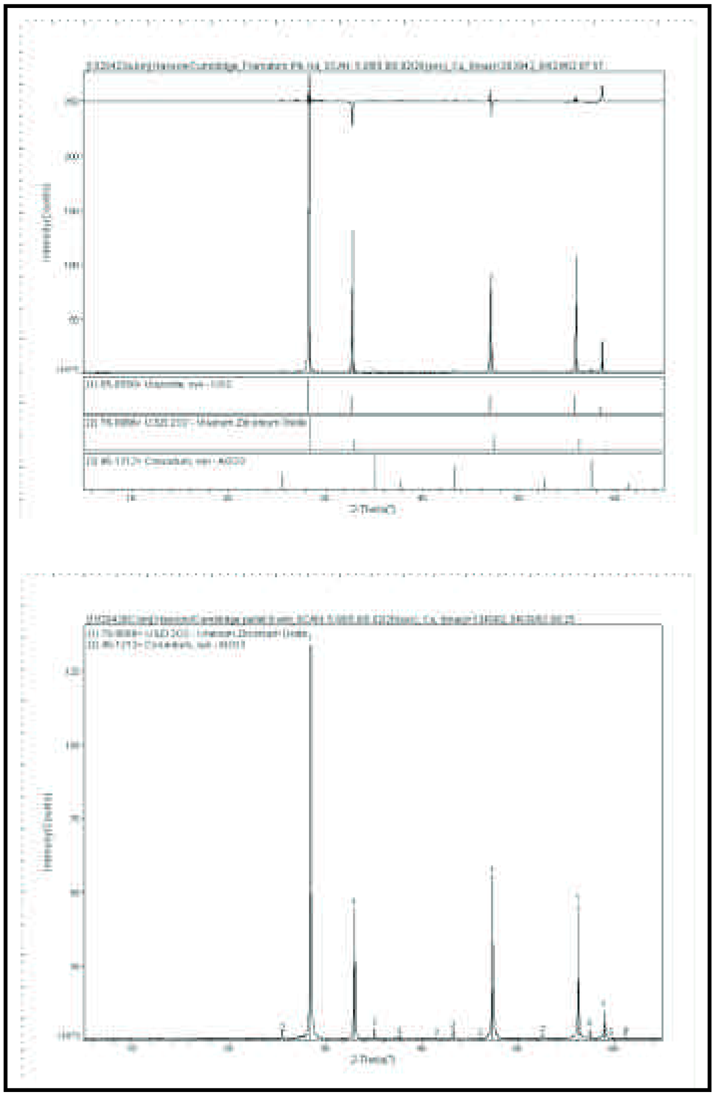


Figure 1. The schematics show XRD fits to 8 wt percent $Gd_2O_3$ (top) and 10 wt percent $ZrO_2$ (bottom) doped $UO_2$ pellets fabricated at PNNL. The Gd pellet is fit using Reitveld refinements of the theoretical lattice parameter with excellent results. The Zr pellet is fit exactly with an established card from the international database. There are no indications of peak broadening or separate phases. Corrundum was added as an internal standard for XRD only.

allow complete development of the HBS formation model.

(2) TGA will be performed on commercial pellets with various Gd-doping levels obtained from Framatome ANP. All pellets produced at PNNL will then be analyzed with TGA to determine the oxidation response. This data will be coupled with the lattice parameters to accurately develop the matrix stabilization model as a function of dopant, ionic radius, and oxidation state so the optimum fuel can be designed.

(3) Additional fuels will be produced and tested for density, oxidation response, and lattice parameter. The thermal conductivities will be measured and the neutronic effects modeled.

(4) Fuels with large grains, both with and without dopants, will be made using high-temperature steam oxidation.

(5) It is hoped that a few candidate fuels will be trace-irradiated in a university research reactor so the fission gas release characteristics can be measured.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Continuous Fiber Ceramic Composite (CFCC) Cladding for Commercial Water Reactor Fuel

Primary Investigator: Herbert Feinroth, Gamma Engineering Corporation

Collaborators: Ceramic Composites Inc.; Massachusetts Institute of Technology (MIT); McDermott Technology, Inc.; Swales Aerospace Corporation

Project Number: 99-224

Project Start Date: August 1999

Project End Date: December 2000

SBIR Project Start Date: August 2001

## Research Objectives

Existing commercial water-cooled nuclear reactors use zircaloy as the reactor structural material for fuel cladding. Zircaloy looses most of its strength over 1,000°F and reacts exothermically with water during low probability events such as loss-of-coolant accidents (LOCAs). As a result, all existing water-cooled reactor systems require complex and redundant safety systems, including rapid acting backup emergency power supplies, to guard against the energy release and hydrogen generation that could occur during a LOCA. The objective of this project is to develop ceramic composite fuel cladding material that behaves at least as well as zircaloy during normal operation, but retains its strength and avoids severe consequences during core overheating accidents such as LOCAs. Consequently, the safety systems of plants that use such cladding could be simplified and made more reliable, leading to more economic plant designs, simplified regulatory requirements, and higher levels of public acceptance.

## Research Progress

In the NERI phase of this project, completed in April 2001, alumina-yttria-based, continuous fiber ceramic composite (CFCC) tubes of about the same diameter as light water reactor fuel cladding, were fabricated and tested in simulated reactor coolant conditions. A 50-day (1,200-hour), in-reactor irradiation and corrosion test in the MIT Research Reactor was conducted at typical pressurized water reactor coolant temperature and chemistry conditions. Results showed that this alumina-based CFCC cladding has good corrosion/erosion resistance under realistic reactor conditions.

Accident tests for simulated loss of coolant were conducted in which representative samples were quenched in water from temperatures ranging from 1,000°F to 2,500°F. The test specimens demonstrated no visible structural damage in the 1,000°F and 1,800°F tests, and the coated specimens showed minor damage, but no major failure, from the 2,500°F tests.

The maximum density achievable on the CFCC specimens, which were fabricated using sol-gel impregnation technology, was about 80 percent of theoretical density. Consequently, these initial specimens were permeable to fission gases, and not acceptable for reactor application.

A follow-on research project was approved by DOE as part of the DOE Small Business Research Initiative, designed to solve the permeability problem. This project, completed in February 2002, developed a hybrid ceramic composite, with an inner layer of high density monolithic silicon carbide, and an outer layer of CFCC impregnated via a Chemical Vapor Infiltration (CVI) process. Silicon carbide was used, rather than oxide based ceramics, because of its excellent high-temperature and thermal-conductivity properties, and because of its ready availability. Several processes were used in fabricating the hybrid structure, including different means of braiding and wrapping the fibers around the monolith in order to achieve good bonding and high strength. Photographs of the resulting product are shown in Figure 1.

All hybrid material specimens were tested and proven to be gas-impermeable up to 125 psi at room temperature. The fabricated specimens showed high matrix material density, significant mechanical strength, and a graceful failure mode when stressed to material fracture (i.e., showing a metal-like stress strain behavior over time).

## Planned Activities

The NERI and SBIR 1 research projects have been completed. It is believed that this hybrid ceramic composite cladding has significant potential for Generation IV reactor concepts that require cladding and structural materials to perform at higher temperatures than can be achieved by using traditional metallic cladding.

NUCLEAR ENERGY RESEARCH INITIATIVE

# An Innovative Ceramic Corrosion Protection System for Zircaloy Cladding

**Primary Investigator:** Ronald H. Baney, University of Florida

**Project Number:** 99-229

**Project Start Date:** July 1999

**Project End Date:** January 2003

## Research Objective

The operational lifetime of light water reactor (LWR) fuel is limited by thermal, chemical, and mechanical constraints associated with the operating conditions of the fuel rod assemblies. A primary limiting factor is the waterside corrosion of the Zircaloy cladding that encases the uranium oxide pellets. Oxidation of the cladding in the high-temperature, high-pressure aqueous environment results in the continuous formation of a zirconium oxide layer that eventually begins to spall, degrading the clad integrity. Protective coatings applied to the Zircaloy cladding can drastically reduce this corrosion and degradation. The objective of this NERI project is to develop ceramic corrosion protection systems for Zircaloy clad for use in LWRs that will allow significantly higher burn-ups resulting in major benefits in plant safety and plant economics. Additionally, the coatings will serve to protect the cladding from fretting failure, a very positive secondary effect.

The major technical challenge for coating a metal with a ceramic protection system is to develop a bond between the low thermal expansion ceramic coating and the more ductile, high thermal expansion metal substrate that will enable the coating to maintain a protective layer even in the subcritical water conditions of a pressurized water reactor (PWR). The stability, interface, and properties of several ceramic coatings on Zircaloy have been examined to select the best combination(s) of coating and processing. Strategies being followed to address this challenge include (1) investigating a range of coating materials including SiC, $Al_2O_3$, and diamond-like carbon (DLC); (2) studying the effect of processing types including chemical vapor deposition (CVD), sol/gel processing, and sputtering, and processing parameters such as substrate temperature; (3) investigating the use of a compliant interlayer to relieve expansion mismatch stresses; and (4) researching the effects of surface topography.

The thickness and composition of the ceramic passivating layers were determined by Auger electron spectroscopy (AES) and X-ray photoemission spectroscopy (XPS). Continuity of the ceramic layers was determined by scanning electron microscopy (SEM). Ceramic coatings were screened for hydrothermal stability by placing samples in an autoclave at 650ºF and 3,000 psi for 24 hours and examining the ceramic coating for continuity by AES and SEM. Electrochemical corrosion resistance was examined by the potentiodynamic polarization (DC method) and electrochemical impedance testing (AC method). Adhesion of the passivating ceramic layers was examined by scratch testing. The scratch-test method consists of the generation of scratches with a spherical stylus (generally Rockwell C diamond, tip radius 200 μm) that is drawn at a constant speed across the coating under either constant or progressive loading.

## Research Progress

Results are presented for each of the three years of the project.

Summary of Results from the First Year

- A study of coefficient of thermal expansion (CTE) data extracted from the literature suggested that $Al_2O_3$ and SiC would be good candidates to match the CTE of zirconium.
- Thermal conductivity considerations indicated that SiC would be a good choice, although $Al_2O_3$ films with less than 50 μm thickness were shown to have only very minor effects upon thermal conductivity and center line fuel temperatures.
- A neutronic analysis indicated that thin ceramic coatings of less than 50 microns would have a negligible effect upon reactivity.
- SiC and carbon were deposited by a plasma assisted chemical vapor deposition (PACVD) process onto

Zircaloy-4 coupons and characterized by Auger electron spectroscopy (AES). The SiC coatings prepared from silacyclobutane (ScB) as the precursor did not have continuity under the initial processing conditions studied. Processing studies were continued in the second year.

- Diamond-like carbon (DLC) coatings were deposited onto Zircaloy-4 coupons by a PACVD process and characterized by AES. The promising results of the first year prompted continuation into the second year.
- $Al_2O_3$ coatings were deposited by an Ultramet proprietary ultraviolet chemical vapor deposition (UVCVD) process onto Zircaloy-4 coupons that had their surfaces modified by either polishing, laser roughening, or chemical roughening. Evaluation of these samples by AES indicated some chloride impurities in the coatings, suggesting a probable point of failure, and so this effort was terminated.
- $Al_2O_3$ coatings were deposited on Zircaloy-4 coupons by sol-gel processing from aluminum alkoxides and characterized by AES. Ultimately, work conducted during the second year showed that no sol/gel coatings would withstand even 24 hours in an autoclave at 650ºF/2,500 psi and so this approach was also abandoned.
- A 6"x4" sheet of Zircaloy-4 was coated with $Al_2O_3$ by a thermal barrier coat (TBC) sputtering process in Praxair's Appleton, Wisconsin, laboratory. The coating was too thick (i.e., 2 mm) and no other coating processor could be identified that could apply thin $Al_2O_3$ by this process, so this approach was terminated.
- Nearly stoichiometric SiC coatings were deposited on Zircaloy-4 coupons by a laser ablation deposition (LAD) process and characterized by AES, X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS). Preliminary economic analysis suggested that this process was prohibitively expensive and the work was not continued in subsequent years.

Summary of Results from the Second Year

- SiC films of approximately 700 nm thickness were deposited by the PACVD process, using ScB and $H_2$ as process gases. XRD, AES, and energy dispersive X-ray (EDX) spectroscopy data showed the film compositions were nearly stoichiometric SiC and uniform, containing little oxygen content. Characterization indicated that film morphologies were influenced by process variables such as substrate temperature during deposition, the ScB/$H_2$ ratio, as well as the surface condition of the substrate. Film thickness, surface morphology, and EDX data indicated good reproducibility in film deposition. Though the results looked promising, Florida's PACVD system proved to be very unstable and reproducibility became very problematic. For this reason, PACVD work was continued at MER Corporation in Tucson, Arizona.
- Sputter-deposited alumina films exhibited very uniform and smooth surface morphologies with sharp interfaces between film and substrate. Problems with poor film adhesion during autoclave testing were partially solved by annealing the films at 500ºC in argon.
- Zicaloy-4 substrates were surface alloyed by first sputtering on a thin layer of aluminum metal. These samples were then oxidized at 500ºC in air for 2 hours and exhibited promising compositional gradient structures. Reducing the aluminum film thickness allowed the oxidation to reach the substrate. The result is that there is no intermediate region of unoxidized metal and the film adhesion properties are improved.
- DLC coatings were deposited onto Zircaloy-4 substrates having various surface finishes by Los Alamos National Laboratory using a plasma source ion implantation (PSII) process. They all exhibited high corrosion resistance and corrosion potential in electrochemical tests.
- Electrochemical corrosion studies showed that surface finish influenced the corrosion behavior of coated Zircaloy-4 substrates. The DLC coated sample with a 600 grit surface finish contained defects that act as initiation sites for breakdown. The DLC coated sample with a 0.3 $\mu$m surface finish provides good corrosion protection during short-term autoclave exposure; however, at longer exposure times all of the DLC coatings failed.
- The sputtered alumina samples completely failed during the electrochemical impedance corrosion testing.
- The PACVD-deposited SiC coatings provided excellent corrosion protection before autoclave exposure. Silicon carbide films with 5 $\mu$m thickness showed significant cracking, which decreased the

effectiveness of these films in protecting the underlying substrate. By comparison, 1 μm thick films exhibited a much larger degree of corrosion protection.

Summary of Third Year Efforts to Date

- Aluminum oxide coatings were prepared by controlled oxidation of aluminum metal films on Zircaloy substrates. Analysis of the oxidized coatings showed that gradient compositions were obtained, with Al, Zr, and O content varying through the coating thickness. X-ray diffraction analysis also showed that a variety of intermetallic and oxide phases (such as $Al_3Zr$, $Al_2Zr_3$, $Al_2O_3$, $ZrO_2$, and $Zr_3O$) were formed in the coatings during processing. None of the alumina coatings tested to date have survived dynamic autoclave exposure (350ºC, 3,000 psi) intact. Well-faceted particulates were observed on the sample surfaces after autoclave treatment, suggesting the hydrothermal re-crystallization to AlOOH.
- A detailed study on the effect of Zircaloy substrate surface treatment on the adhesion of silicon carbide coatings was performed. Substrates were mechanically, chemically, and thermally treated prior to coating deposition to obtain a variety of different surface conditions. Silicon carbide coatings were then deposited on the treated substrates using a PACVD process at MER Corporation (see Figure 1). The adhesion of the resulting coatings was assessed using scratch tests. The results indicated that the surface roughness affects film adhesion, with intermediate surface finishes (substrate polished with 240 grit paper) resulting in higher adhesion than samples with finer (600 grit) or coarser (grit blasted) treatments. None of the silicon carbide coatings tested to date have survived autoclave exposure (350ºC, 3,000 psi).
- DLC coatings (deposited in the last quarter) have not survived autoclave testing (see later explaination), although electrochemical corrosion tests indicate some degree of protection compared to the bare Zircaloy substrates.
- MER Corporation has carried out a detailed economic analysis of the process for coating 1.5 million full-length Zircaloy-4 clad tubes per year with silicon carbide by their PACVD process. Their analysis shows that coating clad tubes with silicon carbide would add an additional cost of $42.00 per tube.
- No processing conditions were found where the ceramic coatings were completely stable in an autoclave, which simulated PWR conditions. A detailed failure analysis was undertaken to determine the mode of failure for each of the ceramic coating types, DLC, SiC and $Al_2O_3$.

Even though very pure single-crystal $Al_2O_3$ showed no observable changes when exposed to the autoclave conditions, alumina coatings produced by surface alloying with sputtered aluminum followed by oxidation or produced by sputtering from a pure $Al_2O_3$ target showed by SEM and glancing angle XRD that significant amounts of boehmite, AlOOH, were produced by hydrothermal growth under these same autoclave conditions (see Figure 2).

The results suggest that the failure of all of the alumina-based coatings under autoclave conditions may have resulted from formation of a less thermodynamically stable form of alumina such as an

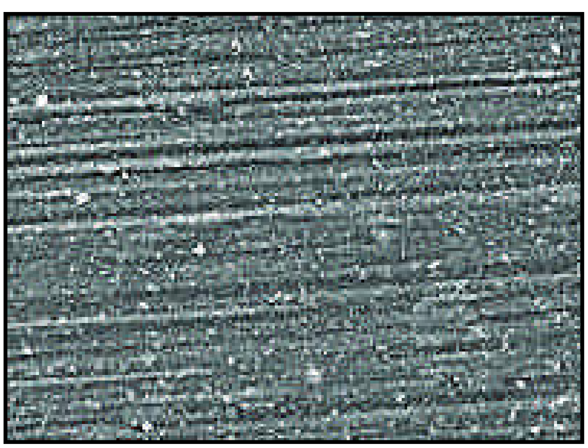


Figure 1. SEM image of the surface of a SiC film (1 mm nominal thickness) deposited on a Zircaloy-4 substrate by PACVD. The coating was deposited at MER Corporation.

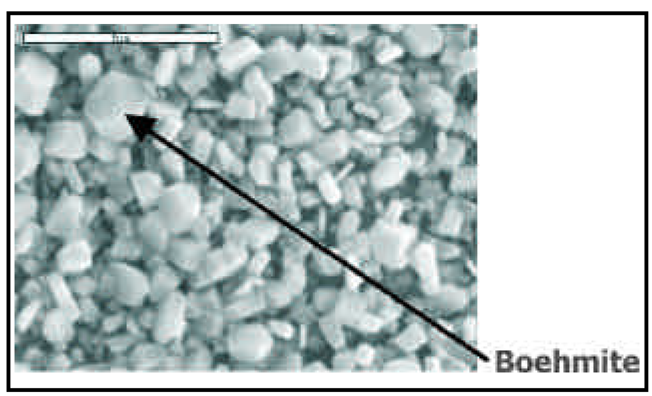



Figure 2. The figure is a SEM micrographs of a 300 nm-thick oxidized aluminum film after autoclave exposure for 24 hours. The aluminum film was deposited on Zircaloy and oxidized at 650°C for 30 minutes followed by 500°C for 2 hours.

amorphous phase or an oxygen deficient phase during the coating process, which would react with water to form the boehmite.

The DLC coatings generally failed after the autoclave exposure by lifting from the surface in large pieces. These pieces would then curl (see figure 3). These results suggest a weak substrate/coating interface but a tough film. The curling after removal from the substrate suggests that the films had residual stresses. The origin of the failure could be a pin hole where the underlying Zircaloy-4 began to oxidize putting stresses onto the film. The DLC appears to be stable, but the bond between DLC and Zircaloy appears to be the problem, with the DLC spalling off.

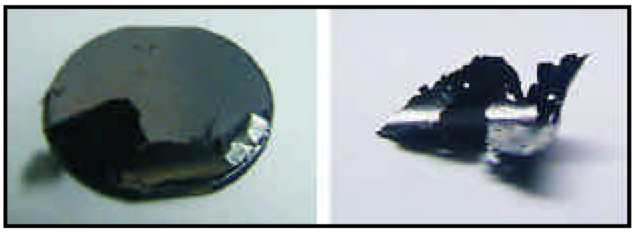


Figure 3. The photographs illustrate the DLC coating after autoclave exposure.

The silicon carbide coatings failed after the autoclave exposure (Figure 4) by first forming blisters generally associated with the the substrate topography caused by the surface finish process before coating. The origin of the blisters is probably pin holes in the coating caused by surface roughness and shadowing effects during the CVD coating process giving rise to holes or thin spots. Oxidation of the underlying zirconium alloy would result in stresses and blistering of the coating.

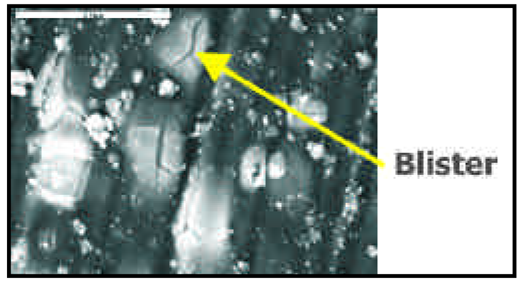



Figure 4. The figure illustrates SEM images of 1 micron SiC film on Zr-4 substrate (600 grit polished), after autoclave test.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Optimization of Heterogeneous Utilization of Thorium in PWRs to Enhance Proliferation Resistance and Reduce Waste

**Primary Investigator:** Michael Todosow, Brookhaven National Laboratory (BNL)

**Collaborators:** Massachusetts Institute of Technology; Ben-Gurion University of the Negev, Israel; the Russian Research Center-Kurchatov Institute (RRC-KI); Commissariat a l'Energie Atomique (CEA), France (inactive); Korea Atomic Energy Institute (KAERI); Kyung Hee University, Korea; Korea Advanced Institute of Science and Technology (KAIST)

**Project Number:** 00-014

**Project Start Date:** September 2000

**Project End Date:** September 2003

## Research Objectives

The objective of this work is to examine heterogeneous core design options for the implementation of the Th-U233 fuel cycle in pressurized water reactors (PWRs) and to identify the core design and fuel management strategies which will maximize the benefits from inclusion of thorium in the fuel. The assessment concentrates on key measures of performance in several important areas including proliferation characteristics of the spent fuel, reliability, safety, cost, environmental impact, and licensing issues. The focus is on once-through fuel cycles that do not involve reprocessing of the spent fuel. A 193 assembly Westinghouse reactor utilizing 17x17 fuel is taken as the model core.

Design optimizations involve heterogeneous core options that aggregate the thorium in subassembly units or whole typical PWR assembly units. The assessment will include comparison to the case of all-uranium fuel (the current fuel cycle and its future extrapolations), as well as the case of Th-U fuel mixtures within individual fuel pins (in both homogeneous and micro-heterogeneous embodiments). Optimization of the homogeneous fuel cycles is being performed under separate projects.

Two heterogeneous thorium implementation options are being explored, and expanded on, in the course of this NERI investigation: 1) the Seed-Blanket Unit (SBU)/Radkowsky Thorium Fuel (RTF) concept, which employs a seed-blanket unit that is a one-for-one replacement for a conventional PWR fuel assembly; and 2) the whole assembly seed and blanket (WASB), where the seed and blanket units each occupy one full-size PWR assembly and the assemblies are arranged in the core in a modified checkerboard array (see Figure 1). The studies for both approaches seek to (1) identify the core design and fuel management strategies that will maximize the benefits from inclusion of thorium in the fuel, and (2) extend the analyses to validate the results over a range of possible operating conditions.

## Research Progress

Designs for both the SBU and WASB approaches have been developed that significantly improve the intrinsic proliferation resistance of the fuel while achieving 18-

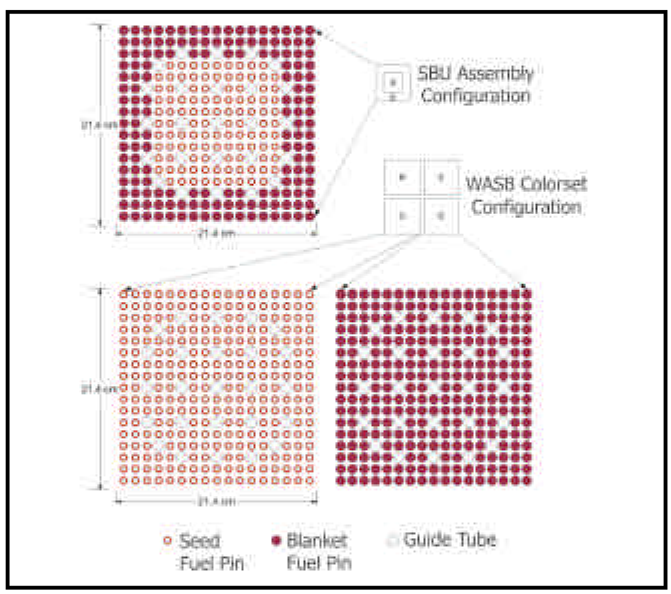



Figure 1. The graphic illustrates various SBU and WASB fuel assembly designs.

month cycles. The total production of plutonium is reduced by a factor of approximately 3-5 relative to a present commercial PWR operating on the uranium cycle. Furthermore, the plutonium that is produced is of inferior quality for potential utilization in weapons: it has a higher heat generation from increased Pu-238, a higher level of radioactivity, and a stronger neutron source from increased Pu-238, Pu-240, and Pu-240. Significantly, both approaches utilize assembly designs based on a Westinghouse 17x17 assembly where the sole modification is in the details of the fuel rods. Therefore, in principle, they are retrofittable into existing PWRs.

In addition, the details of a formalism to evaluate the fuel cycle costs of both SBU and WASB fuel cycles in comparison to a standard PWR cycle have been agreed upon, including the exact formulas and the input data required. The approach includes consideration of the different time periods that seed and blanket components spend in-core, using the levelized fuel cost over a single blanket cycle (typically six to nine operating cycles) as the primary measure of economic value. It also produces comparisons of fuel utilization and material flows. Specific achievements for each approach are summarized below.

The heterogeneous thorium implementation scheme based on the seed-blanket unit is being examined by BNL (with support from Ben Gurion University) under the present NERI project. The project is closely related to an ongoing project supported by the DOE Initiatives for Proliferation Prevention (IPP) program and private funding, which is focused on RD&D activities in Russia and the West. The initial reference point for the optimization studies is based on the results for the PWR from that IPP study.

Progress over the past two years includes the following:

- Conducted independent neutronic and thermal-hydraulic assessment of the reference SBU PWR design. The neutronic evaluations included the MCNP4C and RECOL Monte Carlo codes with explicit geometry modeling and continuous nuclear data, compared to results from the deterministic code BOXER with multi-group cross sections. The results provided initial confirmation of some key aspects of the design.
- Performed simple validation of nuclear data and codes for burnup reactivity and isotopics by analyzing several benchmarks, including the Thorium-Uranium PWR rod-cell benchmark. Results with the BNL methodologies DRAGON and MONTEBURNS (MCNP Monte Carlo + ORIGEN) were in good agreement with results from other codes.
- Defined a reference "improved" SBU design based on sensitivity studies that included varying seed fuel and burnable poison compositions, and rod parameters, within the constraints of a Westinghouse 17x17 assembly design (rod pitch, guide-tube locations). The design assumes annular uranium dioxide pellets with a central burnable poison annulus for the seed, and solid thorium/uranium dioxide pellets for the blanket. The resistance of the grid spacers and the rod outer diameters are profiled to enhance coolant flow into the high-power seed region.
- Adopted a reference mechanical design for the SBU that permits inserting and removing seed rods into an SBU. In the selected approach, rods will be removed and inserted rod-by-rod, using a machine based on one designed to replace failed fuel rods. The virtue of this approach is that the basic mechanical design of the assembly (e.g., grid-spacers, upper and lower end-fittings) can be identical to that currently employed.
- Constructed a detailed COBRA-EN model for the SBU, and performed initial calculations of departure from nucleate boiling ratio (DNBR), and its variation with power. The resultant minimum DNBR based on the well-known W-3 correlation is 1.28 at nominal power based on conservative estimates for pin, assembly, and axial peaking factors, and nominal uniform grid resistances. Therefore, further optimization of power distributions and thermal-hydraulic characteristics is required in order to achieve an acceptable design.

The WASB design assumes that each type of fuel occupies a whole PWR assembly. The assessment of this design is largely performed at MIT.

Progress over the past two years includes the following:

- Designed WASB seed assemblies and blanket assemblies, which, in principle, are backfittable into existing PWRs. They employ oxide fuel in both seed and blanket assemblies (annular uranium dioxide pellets with erbia burnable poison in the central region of the seed, and solid thorium/uranium dioxide pellets in the blanket). For the model Westinghouse PWR, the same 17x17 rod array is utilized as in commercial reactors, but with smaller diameter fuel rods in the seed, and larger diameter rods plus flow-resistance grids in the blanket assemblies, in order to enhance flow in the higher-power seed assemblies and reduce it in the lower-power blanket assemblies.

- Attained acceptable DNB margins under overpower conditions typical of anticipated operational transients through use of these assembly design features, together with the nuclear design of the cores. Evaluations were conducted with a full-core model using EPRI's VIPRE thermal margin code with conservative axial power peaking and thermal-hydraulic inputs.
- Investigated numerous core designs, but the most interesting appears to be one utilizing 84 seed assemblies and 109 blanket assemblies. Its reactivity coefficients are similar to those for commercial PWR cores. Typical three-batch fuel management is employed for the seeds, so that groups of 28 are loaded in sequential cycles and spend three cycles in the core, achieving a discharge burn-up of about 140 MWD/kgU. The 109 blanket assemblies are loaded and discharged together, and spend six to nine cycles in core, achieving a discharge burn-up of about 90 MWD/kgHM.
- Conducted preliminary analyses of fuel rod mechanical behavior at these high burn-ups utilizing the NRCs computer code for this purpose, and identified high fission gas release in the seed and oxide growth on the surface of blanket rods as challenges to the mechanical design of the fuel rods. Subsequent analyses have shown that the use of a larger fuel rod plenum combined with optimization of the burnable poison could probably reduce the fission gas issue in the seed to manageable levels, and that advanced clad types now coming into use, M5 for example, could eliminate the oxide corrosion problem.
- Developed reactor physics methods suitable for "production" analyses of WASB type seed-blanket cores. This will be particularly valuable for future work, since it allows analyses to proceed rapidly using convenient computer codes well-known in the industry. Because the applicability of these codes to WASB type fuel had not been verified, their use has been validated by comparison with Monte Carlo analyses using MCNP and MOCUP. Additionally, the NRC's FRAPCON-3 code for fuel rod mechanical analysis was modified to include both the thermal properties of thorium-uranium oxides, and a calculation of the rim effect in the presence of both U238 and U233.

## Planned Activities

Planned work for the final year of the project will include the following:

- Modify the blanket neutronic design so that the combined total of U233 and U235 does not exceed the proliferation limit proposed by Forsberg at ORNL. Although it is anticipated that this will not be difficult, it will require re-analysis of the core designs to assure that they remain acceptable.
- Evaluate reactivity parameters related to control rods - e.g., total bank worth, stuck rod worth.
- Perform an initial assessment of the safety characteristics of both designs.

NUCLEAR ENERGY RESEARCH INITIATIVE

## High Performance Fuel Design for Next Generation PWRs (Annular Fuel Project)

Primary Investigator: Mujid S. Kazimi, Massachusetts Institute of Technology

Collaborators: Gamma Engineering Corporation; Westinghouse Electric Corporation; Duke Engineering & Services (now Framatome); Atomic Energy of Canada Limited

Project Number: 01-005

Project Start Date: August 2001

Project End Date: September 2004

### Research Objectives

The overall objective of this NERI project is to examine the potential for a high performance advanced fuel for pressurized water reactors (PWRs), which would accommodate a substantial increase of core power density while simultaneously providing larger thermal margins than current PWRs. This advanced fuel will have an annular geometry that allows internal and external coolant flow and heat removal. Details of the tasks follow.

(1) Identify the most promising fuel assembly arrangement of internally and externally cooled annular fuel for PWRs to achieve a significant increase of power density (by at least 30 percent), based, to a large extent, on the extensive PWR fuel database to minimize research and development (R&D) expenses and deployment risks.

(2) Optimize the fuel for superior thermal hydraulic and safety performance. Examine the optimum flow distribution, core pressure drop, maximum departure from nucleate boiling ratio (DNBR), and the resistance against parallel channel instabilities. Perform safety analyses, such as loss of coolant accident (LOCA) analyses, to confirm safety benefits expected for the new fuel.

(3) Evaluate the neutronic fuel design with respect to achievement of high reactivity-limited burnup and reasonably long refueling cycle to attain good economic, waste, and proliferation-resistance features. Confirm that reactivity feedbacks and reactivity control aspects are acceptable.

(4) Select fabrication processes to produce annular fuel elements with the required product characteristics, including fissile loading and high integrity cladding, which are capable of eventual scale-up into a low-cost, efficient production process for economic and reliable fuel element performance.

(5) Evaluate the performance of $UO_2$ fuel forms obtained by production technologies different from current U.S. practices (e.g., vibropacked fuel), and operating under new conditions (especially low peak fuel temperature) on fission gas release, and fuel dimensional properties during burnup. Models will be developed and used for the fuel performance assessment as well as scoping irradiation tests performed at the MIT reactor.

(6) Optimize the core and plant design to minimize the electricity cost in cases of using the annular fuel for uprating current Generation II PWRs or in new advanced PWRs.

### Research Progress

The progress will be summarized according to tasks.

Task 1. Thermal Hydraulic and Mechanical Design and Safety Analysis: A computer code for an isolated channel thermal hydraulic analysis of Internally and eXternally cooled Annular Fuel (IXAF) has been developed and used for the thermal-hydraulic optimization of fuel design. Also, the well-established VIPRE-01 model of the hot fuel assembly having 13x13 annular fuel rods was developed and comparisons made with the isolated channel model. Good Minimum Departure from Nucleate Bioling Ration (MDNBR) agreement was shown between the two models. To identify the optimum dimensions of annular fuel in a square lattice, various array sizes (11x11 to 15x15) that fit in the fixed dimensions of a fuel assembly were explored using the isolated channel model.

The most promising option, based on thermal hydraulic considerations, was found to be a 13x13 array with inner and outer cladding diameters of 8.6 and 15.4 mm, respectively (see Figure 1). This geometry offers the highest Departure from Nucleate Boiling Ratio (DNBR) margin, a low peak fuel temperature and tolerable pressure drop. The 13x13 design, designated as PQN-02, was found to have a peak fuel temperature (at 150 percent power) that is about 1,300°C lower than the reference solid fuel. Therefore, a large power density increase and simultaneous increase of margin for cladding temperature during LOCA is feasible. The major limiting thermal hydraulic parameter appears to be the axial pressure drop across the core, because higher coolant flow rates would be required to maintain a core outlet temperature comparable to current designs and prevent saturated boiling. Additionally, vibration-related issues may limit the core power density increase, since they impose limits on coolant velocity. Therefore, only a 50 percent power density increase was adopted as the target power level. This is a significant power uprate, raising the extracted power from the same core size to support increasing the plant output from the current 1,150 MWe to 1,750 MWe.

In the safety analysis arena, Duke Engineering & Services (DE&S) finished the development of a suitable RELAP5/MOD3.2 base model for a sample PWR. The model simulates a standard Westinghouse 4-loop design, rated at a power level of 3,411 MWth with solid fuel rods and will be used to provide a benchmark calculation. In addition, an evaluation of LOCA performance of the annular fuel in a Standard Korean Nuclear Power Plant (KSNPP) using RELAP5/MOD3.2 was performed at MIT. The peak cladding temperature was found to be ~300°C smaller in comparison with reference KSNPP fuel and 600°C lower in temperature than the Nuclear Regulatory Commission's (NRC's) 1,200°C limit, allowing significant power uprate.

Task 2. Reactor Core Physics and Fuel Management: Initial activities were focused on benchmarking and selection of analysis tools. The MCNP based burnup code MCODE (a recently developed code at MIT) was compared against CASMO4 for a typical unit cell with a $UO_2$ solid pin. A significant discrepancy between CASMO4 and MCODE (Monte Carlo-Origen Depletion) confirmed the expectation that CASMO4 would not be suitable for direct application to IXAF since it employs, as a default, an exponential radial distribution of U-238 resonance integral within a fuel pin. Various options for modifying CASMO-4 input have been investigated to achieve better agreement with MCODE.

The development of a modified CASMO-4 will enable whole core fuel management studies with a standard diffusion code. Using MCNP and MCODE for a unit cell or fuel assembly level, with the same enrichment and core power density, the PQN-02 13x13 design was found to

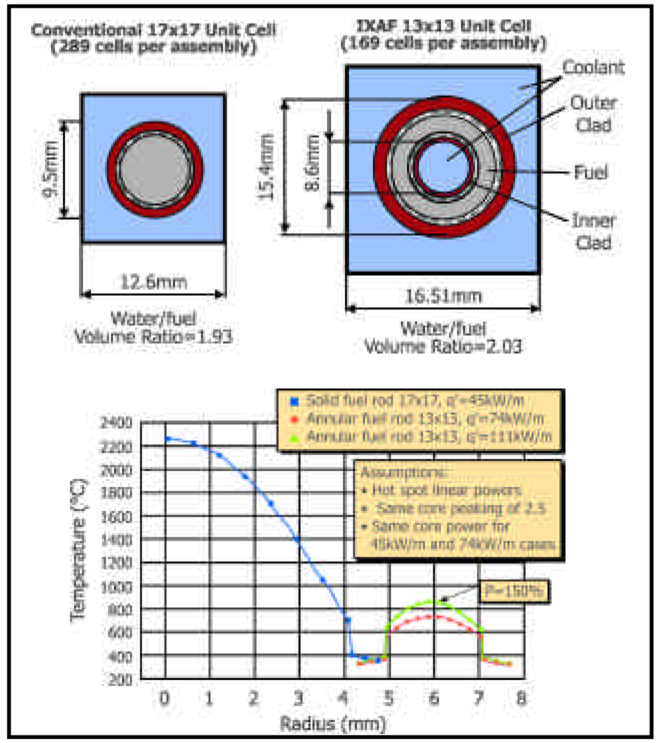



Figure 1. The graphic compares the performance of solid and annular fuel.

achieve the same burnup as the solid pin in spite of larger parasitic losses due to increased cladding volume. This is because of a slightly higher moderator to fuel ratio, higher conversion ratio, and a smaller reactivity penalty from fuel temperature coefficient due to the substantially lower fuel average temperature of the annular fuel. However, less total energy (i.e., fewer effective power days) is achievable because of the smaller heavy metal inventory of the PQN-02 design. Vibro Packed (VIPAC) fuel generally has smaller smear density than the pellet fuel resulting in smaller reactivity-limited burnup than for the pellet fuel. The addition of uranium getter powder, based on Russian manufacturing and irradiation experience, allows the achievement of higher smeared density than in pellet fuel resulting in correspondingly longer cycle. However, the United States does not have experience with nor data for this fuel and an R&D program if required.

Task 3. Fuel Fabrication Studies: Analyses of key design and performance requirements of optimized annular fuel elements were conducted at Gamma Engineering, with particular focus on those requirements that affect the selection of annular fuel fabrication technology. Potential fabrication routes and processing technologies of annular fuel elements were identified as the following:

(1) Sintered ring pellets fabrication route with traditional punch and die technology for fabricating green pellets before sintering processing;

(2) Sintered ring pellets fabrication route with slurry extrusion technology for fabricating green pellets before sintering processing;

(3) Sintered ring pellets fabrication route with tape casting technology for fabricating segments of green ring pellets before sintering processing;

(4) VIPAC fuel element fabrication route with different particle size components of crushed high density sintered $UO_2$ fuel material; and

(5) VIPAC fuel element fabrication route with different particle size components of spherical high density $UO_2$ fuel material formed by special sol gel processes.

A subcontract to Atomic Energy of Canada, Ltd (AECL) for the fabrication of VIPAC annular fuel test specimens for irradiation at MIT reactor has been signed and became effective in April of 2002. The delivery of finished products to MIT for in-reactor testing is scheduled in November 2002.

<u>Task 4. Economic Analyses and Optimization:</u> For manufacturing cost evaluations, the baseline constraints and assumptions adopted were those that reflect the current permitting and operational constraints at the Westinghouse Nuclear Fuels plant in Columbia, South Carolina. Changes for all manufacturing procedures associated with annular fuel have been identified and the cost difference is under evaluation. A preliminary assessment indicates small increases in manufacturing costs are expected, about 0.5mills/KWhe.

For the fuel cycle and capital cost assessment, an approach was developed for the evaluation of the costs and benefits of the high-power enabled by the annular fuel and preliminary analyses were performed. The early results indicate that the benefits of increasing the power density in the vessel of new PWRs easily exceed the incremental costs associated with fuel manufacturing.

<u>Task 5. Fuel Performance Evaluation:</u> The activities focused on preparing the design of the fuel irradiation experiment and on fuel modeling. The design of the test specimens have been proposed and analyzed to ensure that temperature profiles, representative of those in the power reactor, can be achieved under sufficient cooling at the MIT Research Reactor (MITR). Application for license amendment has been submitted to the NRC. NRC's questions have been answered and minor revisions are currently being completed. The Safety Evaluation Report for approval of the planned experiment by the MITR-II Reactor Safeguards Committee is nearing completion and the draft is currently being reviewed by the MITR-II Operations staff.

For the fuel performance modeling, the FRAPCON-3 code is being modified for IXAF. The structure of the original FRAPCON-3 has been redesigned and the proper code initialization has been established for the annular fuel. Calculations of inner channel coolant conditions, cladding corrosion, crud uptake, hydrogen concentration, film temperature drop, and temperature drops in crud, oxide layer, and across inner cladding have been performed. The gas production and burnup subroutines have also been modified for annular conditions. A new fuel cladding mechanical interaction model, based on the existing FRACAS-I model, has been formulated and tested for annular fuel.

## Planned Activities

Future activities will be focused primarily on the following efforts:

Task 1. The development of a VIPRE whole core model to obtain more accurate DNBR analysis, and study of the impact of geometrical uncertainties and partial blockage of the inner channel on thermal hydraulic performance. Additionally, assessment of mechanical design issues, in particular flow-induced vibrations and stability against liftoff.

Task 2. Calculation of reactivity feedbacks and boron and control rod worth, design of control strategy and whole core model for fuel management, and further optimization to achieve 12-month cycle length at 150 percent power.

Task 3. The manufacturing of annular fuel elements, using natural uranium or surrogate as the fuel body, using one, or possibly two of the processes examined in year 1. Varying of fabrication processing parameters to determine those parameters that offer the best promise of meeting the fuel product and performance requirements.

Task 4. The quantitative determination of marginal fabrication cost of annular fuel for sintered pellets and vibropack fuel, and evaluation of the economic benefit to PWRs from capital and fuel cycle costs using annular fuel at up to 150 percent power density.

Task 5. The completion of the FRAPCON-3 computer code for analysis of annular fuel performance for both pellet and VIPAC fuels, and of the design work for irradiation of VIPAC test specimens to be loaded in the MIT reactor in January 2003.

NUCLEAR ENERGY RESEARCH INITIATIVE

# An Innovative Transport Theory Method for Efficient Design, Analysis, and Monitoring of Generation IV Reactor Cores

Primary Investigator: Farzad Rahnema, Georgia Tech Research Corporation

Collaborators: Pennsylvania State University; Idaho National Engineering & Environmental Laboratory

Project Number: 02-081

Project Start Date: September 2002

Project End Date: September 2005

The current generation of core neutronics methods are based on nodal diffusion theory and utilize homogenized cross sections and other physics data generated by single assembly, infinite medium transport theory calculations. This reactor-analysis methodology was developed and refined for the currently operating class (Generation II) of light water reactors (LWRs). Until about a decade ago, the reload cores of these reactors were designed with relatively homogeneous distributions of fuel, moderator, and absorber materials. For these systems, core-level diffusion theory is a good approximation, and the computational de-coupling of fuel assemblies for generating physics data is acceptable.

The current trend in LWR cores, however, is toward higher degrees of heterogeneity. In order to lengthen operating cycles, recent cores have been designed with higher amounts of total fissile mass, which has necessitated the addition of burnable absorbers to hold down the reactivity at the beginning of core life. Increased fuel utilization has been achieved by varying the fuel enrichment within assemblies and optimizing the arrangement of assemblies with significantly different fissile and fission product compositions. It is reasonable to expect these or similar design features to be present in the Generation IV light water reactors due to the unchanging desire to increase plant availability and reduce cost.

The trend toward compositional heterogeneity in LWRs and the desire for smaller, modular reactors in the Generation IV class will lead to cores with higher neutron flux gradients, resulting from increased core surface area to volume ratios in the latter case. In these systems, the neutron leakage between adjacent assemblies is significant and cannot be neglected. Generating physics data using single assembly infinite medium transport calculations, as is done with current core neutronics methods, may lead to substantial errors in the homogenized cross sections and discontinuity factors. Without accurate data, the simplified nodal core model will produce inaccurate results. This is the consequence of the computational de-coupling of a highly coupled system.

Non-LWR Generation IV reactor designs are likely to be so different from current LWRs that they will necessitate a different (and probably smaller) set of assumptions on which to base core neutronics models. For example, in the pebble-bed modular reactor (PBMR), a high degree of uncertainty exists in the distribution of fissile mass among localized core regions due to the movement of pebbles with different degrees of burn-up as well as the presence of pebbles that contain only graphite. Further uncertainties resulting from the use of computational methods based on approximations to transport theory with limited ranges of validity will only exacerbate this problem. In addition, accurate calculations in localized portions of the PBMR cores must be performed in three spatial dimensions due to the complex geometry of packed arrangements of spherical pebbles. This aspect of PBMR cores creates problems for the current methods based on two-dimensional transport calculations applied to LWRs in which the variation of core properties in the vertical (third) dimension is relatively weak.

It is clear that the next generation of reactor analysis methods will be based largely on transport theory (both at the assembly and core levels) and involve fewer approximations regarding the nature of the core system than current methods. Diffusion theory and the multitude of methods based on transport corrections to diffusion theory will not be sufficient to support the optimum design, operation, and monitoring of the next two generations of reactor systems for the reasons delineated above. A computationally efficient core-wide transport theory method would provide a highly accurate and flexible design tool (i.e., applicable to a much broader class of systems). In addition, from an engineering standpoint, it would support the pursuit of maximal increases in fuel utilization and plant availability and decreases in operating margins, the probability of fuel damage, and spent fuel inventory. These are many of the advantages sought in the Generation IV class—all of which lead to reduced overall costs.

The currently available transport theory methods have had limited success when applied to core-level calculations, and nearly all require the homogenization of assembly-level physics data. It is proposed that a next-generation, high-order variational coarse-mesh transport method be developed that does not require any homogenization or the use of discontinuity factors. The method is developed by deriving equations from a variational principle that admits discontinuous trial functions. Surface Green's functions are used for the spatial basis within each coarse-mesh and include all of the local transport characteristics as opposed to polynomial or other simple basis sets. Preliminary work in one-dimensional slab geometry with discrete ordinates and multigroup cross sections has been completed and demonstrates the feasibility and promise of the approach. The fine-mesh results are reproduced exactly by the coarse-mesh method in the test problems. Integral to the proposed method, and therefore requiring no additional development, is the procedure for flux reconstruction at the detail of the fine-mesh calculations. The speed of the core calculation in higher dimensional geometries is expected to be close to that of current methods so that it can be used for design and core monitoring calculations.

The objective of the project is to develop the transport theory method and implement it for advanced and Generation IV LWRs and for the PBMR. The work will be accomplished through the collaborative effort of three organizations: Georgia Institute of Technology, Idaho National Engineering and Environmental Laboratory (INEEL), and Oak Ridge National Laboratory (ORNL). Georgia Tech will lead the project, develop the transport method, and implement the method for LWR calculations; INEEL will provide expertise in the area of PBMR calculations, and couple a coarse-mesh computational module to their pebble transport (movement) code; and ORNL will provide expertise in performing efficient and accurate transport calculations for both reactor types.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Advanced Extraction Methods for Actinide/Lanthanide Separations

Primary Investigator: Michael Scott, University of Florida

Collaborators: Argonne National Laboratory (ANL)

Project Number: 02-098

Project Start Date: September 2002

Project End Date: September 2005

The separation of An(III) ions from chemically similar Ln(III) ions is perhaps one of the most difficult problems encountered during the processing of acidic nitrate nuclear waste. In the 3+ oxidation states, the metal ions have an identical charge and roughly the same ionic radius. They differ strictly in the relative energies of their f- and d-orbitals, and in order to separate these metal ions, ligands will need to be developed that take advantage of this small but important distinction. If an efficient protocol can be developed for their partitioning, neutron bombardment can be employed to transmutate actinides into products with significantly shorter radioactive lifetimes. Along with aiding the processing of acid nuclear waste streams, this methodology should ease some of the concerns expressed by the American public involving the long-term storage of nuclear waste.

With the intent to mimic the 3:1 CMPO:actinide stoichiometry of the extracted species in the TRUEX nuclear waste treatment process, a ligand system containing three pre-organized carbamoylmethylphosphine oxide (CMPO) moieties anchored onto a rigid three-fold symmetric triphenoxymethane platform has been developed for facile complexation of actinide ions and subsequent extraction from acidic nitrate nuclear waste streams. The CMPO arms on the ligands are oriented such that all three CMPO moieties can cooperatively bind a metal ion. Preliminary extraction experiments with simulated nuclear waste streams with solutions of the first generation of the ligand reveal a high affinity for the actinide thorium and a very low, constant affinity for the lanthanides across the series. Several different ligand derivatives have been prepared, and a series of distribution ratio measurements will be performed at Argonne National Laboratory with at least two lanthanides and americium to test the selectivity of the ligands for the 3+ metals. This information will be used to help design an improved ligand set.

One method to tune the actinide selectivity will be to influence the charge density of the metal and its coordination geometry. Accordingly, small alterations can be made to the ligand system to exploit these differences and further increase its affinity for actinides. Procedures have been outlined to incorporate modifications that alter the basicity of the CMPO oxygen donors as well as the distance between adjacent CMPO groups on the triphenoxymethane platform. With a wide variety of methods to alter the three arms, the binding attributes of the ligand can be subtly adjusted using the extraction data obtained from the group at Argonne to maximize the selectivity for the 3+ actinides.

The proposed work falls within the scope of fundamental chemistry program in the Nuclear Energy Research Initiative and funding from the Department of Energy will allow for a detailed examination of the An(III) binding properties of the ligands at Argonne National Laboratory. This information will be crucial for the further refinement of the C3-symmetric actinide binder at the University of Florida. The continued cooperation between the two organizations should produce an advanced extraction process for the separation of the chemically similar actinides and lanthanides found in acidic nitrate nuclear waste streams. It is envisioned that the compounds prepared during this work can be used in a process to remove the An(III) ions from nuclear waste streams immediately following the PUREX process and perhaps after concentration of the An(III) and Ln(III) ions by the DIAMEX process.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Improving the Integrity of Coated Fuel Particles: Measurement of Constituent Properties of SiC and ZrC, Effects of Irradiation, and Modeling

Primary Investigator: Lance L. Snead, Oak Ridge National Laboratory

Collaborators: CEGA Corporation, Idaho National Engineering & Environmental Laboratory; Massachusetts Institute of Technology; General Atomics; Argonne National Laboratory

Project Number: 02-131

Project Start Date: September 2002

Project End Date: September 2005

The silicon carbide (SiC) layer integrity in the TRISO-coated gas-reactor fuel particle is critical to the performance, allowed burn-up, and hence, the intrinsic efficiency of high-temperature gas cooled reactors. While there has been significant developmental work on manufacturing the fuel particles, the effects of the complex in-service stress state combined with realistic materials property data under irradiation has on fuel particle survival are not adequately understood. Furthermore, there is virtually no experimental data on the effects of irradiation on the thermo-mechanical properties of zirconium carbide, which has been proposed as a higher-temperature replacement for SiC. The basic assertion behind this proposal is that a significant need exists for detailed fuel particle modeling including realistic, experimentally derived data on fuel particle constituent materials in the non-irradiated and irradiated condition. To perform this work will require advances in modeling, along with development of techniques for measuring materials properties at the small scale of the fuel particle.

Four elements are proposed for this work:

(1) Modeling Work: In recent years, a collaboration has been established between Idaho National Engineering and Environmental Laboratory (INEEL) and the Massachusetts Institute of Technology (MIT) to look into finite-element and other methods of modeling the stress state of fuel pellets. This work has been carried to the point where it is being limited by the lack of realistic material property input. Specific information on the statistical distribution of strength, creep, and swelling for SiC is poorly described in the literature. Data on the thermomechanical properties of pyrolytic ZrC is also very limited. This new data will be generated and applied in spherical and cylindrical model geometry. The objective of this work is to use the new data to better describe the stress state of the TRISO particle under irradiation and to give a direct comparison of the integrity of SiC-v-ZrC for this application. Potential failure during pellet processing will also be addressed.

(2) Technique Development for Measuring Constituent Properties: To this point, techniques to study the integrity of fuel particles have been relatively rudimentary, consisting of compression tests (crush or c-ring) of the particle or bare SiC overcoat. The objective of this element is to apply state-of-the-art techniques and to develop new techniques specifically for application to the TRISO system to generate realistic data for the modeling. These techniques would then become available for the community developing gas-cooled-reactor fuels. Specific tools will be developed to measure strength through internal pressurization, elastic moulus on the scale of the TRISO particle, creep relaxation, and PyC/SiC interfacial properties.

(3) Irradiated Materials Property Information: An irradiation program will be coupled with the technique-development program to generate information on mechanical properties needed for modeling input. The irradiation program will include the model, non-fueled cylindrical and spherical TRISO structures, and spherical TRISO-containing, helium-producing boron carbide.

(4) Updated Materials Data Handbook for TRISO Fuels: As part of this effort, a materials property

handbook will be developed. This handbook will include pertinent physical property information on all constituent materials of coated particle fuel. Sources of information will be the open literature on nuclear materials, reports dealing with HTGRs (e.g., CEGA-002820, Rev 1), and information developed as part of this proposal. This material will then be available to the larger fuels community.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Enhanced Thermal Conductivity Oxide Fuels

Primary Investigator: Alvin Solomon, Purdue University

Collaborators: Framatome ANP, Inc.

Project Number: 02-180

Project Start Date: September 2002

Project End Date: September 2005

The objective of the proposed research is to produce a novel oxide fuel form with superior thermal conductivity. The research is proposed under NERI's Materials Science area of emphasis and enhances the long-term viability and safety of nuclear energy systems and the stability of the spent fuel as a waste form. The resulting fuel will be applicable to existing light-water reactors, especially with high burn-up, high performance fuels. It is also expected that such fuel will provide superior performance in advanced reactors that would otherwise be fueled with low-conductivity oxide fuels.

Although $UO_2$ fuel has many desirable chemical characteristics and has served satisfactorily in light water reactors for many years, its low thermal conductivity imposes significant limitations on reactor operations in present and especially in high performance, high burn-up future reactors. An increase in the thermal conductivity would relax several of these limitations and provide the following significant benefits:

(1) Operational safety would be enhanced because fuel performance during a LOCA (loss-of-coolant accident) would be improved. The amount of heat stored in the fuel would be reduced, so peak cladding temperatures during dryout would be reduced.

(2) A reduction in fuel temperatures and stored heat would support reactor power up-rates and improve power production economics.

(3) The production of high-level radioactive waste would be reduced because fuel burn-up could be increased since lower fuel temperatures would result in less fission gas release, and smaller amounts of fission gas could eliminate limitations on burn-up that are tied to internal pressure of the fuel rod.

(4) Reduced temperatures and temperature gradients in the fuel pellets would reduce the stresses imposed on the cladding, reduce fuel cracking and relocation, and reduce life-limiting fuel swelling, so the effects of pellet-cladding mechanical interaction would be reduced.

(5) Proliferation resistance would be enhanced because the rate of production of $^{239}Pu$ decreases as burn-up increases.

The goal of enhancing the thermal conductivity of sintered oxide fuels will be achieved by a new process of penetrating a highly conductive solid second phase into the open or interconnected porosity of sintered fuel. This project would focus on $UO_2$ for the present program because the technology can be immediately applied to current LWR fuels.

The process first involves developing sintering schedules to produce a desired open pore structure (approximately 90 percent TD). The second critical step requires intrusion into the open porosity of the high conductivity phase. Although penetration of the porosity with a molten metal under pressure is conceivable, this requires very high processing temperatures for high-melting-point refractory metals or even zirconium alloys. A new, more attractive alternative method would be to infiltrate a liquid silicon carbide (SiC) polymer precursor and pyrolyze it at modest temperatures leading to an interconnected SiC phase with superior thermal conductivity for fully-dense polycrystalline material comparable to that of commercial purity silver (approximately 400 W/m-°K at room temperature). Although several infiltrations and pyrolyzing steps may be necessary, they could be readily performed in a simple batch process. Thus, it is proposed that the second approach be pursued.

Besides high thermal conductivity, necessary characteristics of a second phase include small neutron capture cross sections; high melting point; resistance to creep; compatible thermal expansion coefficients; and lack of chemical reactions with $UO_2$, fuel cladding, or water. SiC has been identified as an excellent material from most of these perspectives. For example, its thermal conductivity at 1,000°K is roughly ten times that of $UO_2$, and improves at higher temperatures. Moreover, the methodology for deposition of SiC carbide has been developed.

Standard $UO_2$ fuel has a very large temperature drop between the center and surface of the fuel pellets during irradiation. A drop of 1,000°C is not unusual during full power operation. The primary reason for the large temperature drop is the low thermal conductivity of $UO_2$. Substantial increases in thermal conductivity are possible with modest volume loadings of the conductive phase. Preliminary model calculations indicate that if the second phase has a high thermal conductivity compared to $UO_2$, a 10 percent volume loading of a continuous, randomly oriented second phase would increase the thermal conductivity of the fuel by about 50 to 100 percent, depending on temperature, and reduce the peak centerline fuel temperature in the hot channel bundle with a linear power of 29.8 kW/m by 800°C. It would be even greater for the peak pin.

The first technical challenge in producing an enhanced-conductivity fuel is first to model the heat-conducting performance of various microstructures. Three aspects of the microstructure are essential to good fuel thermal performance. First, the second phase must be finely dispersed. Widely spaced conductive paths would be less effective in conducting heat from the fuel and could produce localized hot spots on the cladding, resulting in nonuniform corrosion and hydrogen migration in the cladding. On the other hand, if the porosity is too fine and finely dispersed, penetration becomes difficult and thermal conductivity may be reduced by phonon scattering in the conductive phase. Second, the conductive phase must be continuous. Although there is a slight improvement of heat transfer even from discrete particles of a highly conductive material, much larger benefits are derived if the conductive material forms a continuous path. Third, the conductive network must penetrate the entire fuel pellet so that heat can be conducted efficiently from the center to the surface. Recent advances in SiC processing make it possible to produce SiC that has a very high thermal conductivity because of its high purity and good crystallinity. Doping with appropriate n-type elements can further increase the conductivity to above 480 W/m-°K. Silicon carbide also has a melting point of 2,700°C consistent with that of $UO_2$.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Use of Solid Hydride Fuel for Improved Long-Life LWR Core Designs

Primary Investigator: Ehud Greenspan, University of California

Collaborators: Massachusetts Institute of Technology; Westinghouse Savannah River Company; University of Tokyo

Project Number: 02-189

Project Start Date: September 2002

Project End Date: September 2005

The general objective of this proposal is to assess the feasibility of improving the performance of PWR and BWR cores by using solid hydride fuels or solid hydride inserts. The concentration of hydrogen in the hydride fuel is comparable to that of hydrogen in liquid water of LWR cores. The introduction of part of the hydrogen needed for neutron moderation within the fuel volume permits attainment of optimal neutron spectrum while using a relatively small water volume fraction-just that amount of water that is required for comfortably safe cooling of the fuel. This feature enables the core to be designed to have optimal moderation, in terms of the attainable discharge burn-up, and to have a smaller volume or higher total power than a LWR core that uses oxide fuel. Moreover, thorium hydride fuel, one of the hydride materials to be examined, has a higher heavy metal (HM) density than oxide fuel. As a result of this higher HM concentration and larger fuel-to-water volume ratio, U-$ThH_2$ or Pu-$ThH_2$ fueled cores can be designed to have a significantly higher energy generation per core loading and significantly longer core life than the corresponding oxide-fueled cores. Preliminary estimates indicate that both the energy per fuel loading and core life could increase by more than a factor of 2. The core power level can also be significantly increased. The net outcome is expected to be improved economics, improved resource utilization, reduced waste, improved proliferation resistance, and improved safety.

This study may lead to the development of new fuel and core designs for LWRs that could have one or a combination of the following advantages relative to contemporary LWRs and those under development:

(a) Reduced capital cost by virtue of compaction and/or increased power output;

(b) increased discharge burn-up;

(c) increased core-life;

(d) increased energy generation per fuel loading;

(e) reduced fuel cycle cost;

(f) reduced waste volume and toxicity due to higher discharge burn-up and to partial utilization of Th;

(g) increased utilization of Pu relative to MOX fueled cores due to the higher discharge burn-up possible with hydride fueled cores of acceptable power density;

(h) utilization of thorium fuel resources;

(i) simplified design of BWR fuel assemblies and control systems along with improved stability against power oscillations;

(j) improved safety due to the large negative temperature coefficient of reactivity of hydride fuel;

(k) improved capability to dispose of plutonium in LWRs by using fertile-free $PuH_2$ or Pu-Zr hydride fuel; (the large prompt negative temperature coefficient of reactivity of hydride fuel may compensate for the lack of large negative Doppler reactivity effect due to absence of fertile fuel); and

(l) improved proliferation resistance due to enhanced destruction of Pu and use of thorium.

One hydride fuel being considered is uranium-zirconium hydride, similar to that developed by General Atomics (GA) for TRIGA reactors. This fuel has been in use for more than 40 years in many reactors around the world, both in constant power and pulsed power operating conditions. It has been extensively studied, and tested in reactors, and it has an impressive record of safety. Fuel for high power TRIGA reactors has been operating under conditions that in many aspects meet or exceed LWR fuel performance requirements. Relative to $UO_2$ fuel in LWRs, this TRIGA fuel operates at close to twice the average linear-heat-rate, and reaches more than twice the discharge burn-up. The thermal conductivity of the TRIGA fuel is nearly six times larger than that of $UO_2$ so that its peak fuel temperature under typical LWR operating conditions is estimated to be below 700°C, which is acceptable and provides a comfortable margin. Some of the novel core designs with hydride fuel being proposed will feature lower linear heat rate and hence, lower peak fuel temperatures. This will result in a large margin to accommodate transients that lead to fuel temperature increase. Uranium-zirconium hydride fuel was also used in a sodium-cooled SNAP space power reactor developed by Atomics International.

Another hydride fuel proposed for consideration is uranium-thorium hydride. It was proposed as fuel for nuclear reactors by the late Dr. Masoud Simnad, the developer of the TRIGA fuel. $U\text{-}ThH_2$ is even more stable than U-ZrH1.6 fuel and can operate at higher temperatures. More importantly, the HM density in the $U\text{-}ThH_2$ fuel can exceed the U density in $UO_2$. It is estimated that by using U-ThH2, it is possible to load more than twice the amount of HM into a core of a given volume than in the corresponding well- moderated $UO_2$ (or MOX) fueled core. This implies that it might be possible to extend the time between refueling by more than a factor of two, or, in principle, double the core power level.

Prof. M. Yamawaki of the University of Tokyo has recently developed and tested a U-Th-Zr-H fuel and carried-out performance tests including in-core irradiation. The maximum permissible linear heat rate limit of that hydride fuel is estimated to be 500 w/cm, more than is needed for economic LWR operation. Prof. Yamawaki will collaborate on this study and provide needed data. Before passing away several months ago, Dr. Simnad had also expressed great interest in participating in this proposed project.

Another, smaller part of the study will assess the feasibility of designing PWR and BWR cores to have long life and large discharge burn-up using very compact lattices incorporating small amount of non-fuel containing solid hydride. The function of the solid hydride is to limit the spectrum hardening upon 100 percent voiding of the water coolant, and thereby help achieve a negative void coefficient.

The study will address primarily reactor physics, and thermal-hydraulic, safety, fuel-cycle, and economic considerations. Material compatibility research will be undertaken as a follow-on study provided the conclusions from the present study justify doing so. To ensure that the present study will adequately take into account material-compatibility issues, two material specialists, Prof. Olander of the University of California at Berkeley, and Prof. Yamawaki of the University of Tokyo, an expert on hydride fuels, have been included on the team.
A complete list of the collaborators, their institutions, and their areas of expertise follows:

(1) Greenspan , University of California at Berkeley, neutronics

(2) Olander, University of California at Berkeley, material compatibility

(3) Todreas, Massachusetts Institute of Technology, thermo-hydraulics and safety

(4) Petrovic and Garkisch, Westinghouse, practical fuel and core design considerations and economics

(5) Yamawaki, University of Tokyo, hydride properties and compatibility

The funding for Prof. Yamawaki's involvement will be provided by Japanese sources.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Development of Advanced Methods for Pebble-Bed Reactor Neutronics: Design, Analysis, and Fuel Cycle Optimization

Primary Investigator: A.M. Ougouag, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Georgia Institute of Technology; Pennsylvania State University; Imperial College of London, PBMR (Pty) Ltd.; University of Arizona

Project Number: 02-195

Project Start Date: September 2002

Project End Date: September 2005

The goal of this project is to develop a comprehensive suite of computer codes for the design of pebble-bed reactors (PBRs) and the management of their fuel cycles. The PBR concept is a leading candidate for near-term deployment and for further development as a Generation IV reactor, yet the neutronics methods available to design and analyze PBRs are several generations behind the state of the art. Existing PBR analysis codes use finite-difference or statistical methods, which are slow and thus unsuitable for the repetitive simulations needed for optimization. Thus, an efficient deterministic method is needed for design and optimization of the PBR fuel cycle.

The INEEL has developed a new deterministic method for the neutronics analysis of PBRs. The method accounts for the pebble flow explicitly and couples the flow to the neutronics. It can model once-through cycles and cycles that allow recirculation of pebbles through the core. The method is implemented in the INEEL code PEBBED. At present, PEBBED uses a finite-difference neutronics solver and simplistic depletion and cross-section computation techniques; these methods were applied to prove the viability of the basic algorithm in the code, but they should be replaced by modern techniques that fully complement the advance achieved by that algorithm.

The project will incorporate the needed modern techniques; beyond that, it will extend the state of the art in computational neutronics. A new method will be developed to carry out long depletion steps while maintaining high fidelity in the model nuclear data. The method will be an essential tool for the reliable design of a PBR or any reactor core of cylindrical geometry. No method or code exists that contains the capabilities proposed here.

The new method and tools will greatly enhance the scientific and computing infrastructure in the United States. The resulting codes will be essential to cost-effective PBR design, and they will give unprecedented fidelity to PBR modeling. Thus, the project will directly support two areas of the NERI work scope: F-1, Advanced nuclear energy systems, and F-3, Nuclear fuels/fuel systems. The project conforms to the NERAC Long-Term Nuclear Technology R&D Plan in advancing reactor physics technology for a Generation IV reactor concept, and it will enable assessment of the proliferation potential of PBRs as advocated by the NERAC TOPS report. It is also consistent with the National Energy Policy of 2001, which specifically mentions the PBR as a promising advanced reactor concept.

The following paragraphs explain the specific needs addressed in this project.

<u>Implementation of a 3-D (r-θ-z) nodal/coarse mesh multigroup neutron diffusion solver</u>: A way has been found to circumvent the mathematical impasse that has frustrated all previous efforts to find a mathematically rigorous formulation of an analytical nodal method in 3-D cylindrical geometry. In real PBRs, the presence of control rods or control elements, uneven burn-up, and uneven build-up of poisons will impose azimuthal asymmetry (i.e., θ-dependence). To perform core physics analyses efficiently in most reactor systems, nodal methods are generally applied. However, current nodal methods in cylindrical geometry do not include the θ-dependence rigorously because the transverse integration procedure (from which nodal method developments start) in r leads to a mathematical impasse. In this project, a new mathematically rigorous approach will be implemented into a nodal code for r-θ-z geometry.

Development of an advanced, spatially detailed depletion capability: A new nodal depletion method will be developed in this work, extending to cylindrical geometry the techniques in the NOMAD-BC code developed by one of the PIs,. The current standard methods used for PBR design and analysis rely on finite-difference neutronics solvers. Consequently, they are limited either to very low-fidelity modeling (by a very coarse computational grid) or to very long computational times (if the computational mesh is refined). Such methods in detailed design and safety studies would impose an unacceptable bottleneck by modern standards. The nodal/coarse-mesh-based depletion capability proposed in this project would eliminate this difficulty, because nodal methods have the potential to increase fidelity while greatly reducing computational times. With the new method, complete design analyses could be completed in a matter of seconds. Optimization studies would also be commensurately rapid.

Development of a new method for generation of diffusion-theory parameters: The nodal diffusion solver and nodal depletion method will be complemented by a cross-section perturbation method that will be developed in collaboration with Prof. Farzad Rahnema and his students at the Georgia Institute of Technology. This method will be an extension to cylindrical geometry of prior work at Georgia Tech. The method will be supported by the EVENT code developed by Prof. Cassiano de Oliveira at the Imperial College of London; Prof. Oliveira will collaborate as an unfunded participant. The new method extends modern equivalence theory past the present state of its art. Besides homogenized diffusion constants, the method will produce perturbation parameters, used to update nuclear data during changes in the reactor core that develop during long time steps, without resort to data-intensive, pre-computed tables or rehomogenization. This capability is unique and extremely well-suited to first-of-a-kind reactor design and analyses; it is also ideal for efficiency and fidelity in the analysis of existing reactor types.

Development of a method of feedback parametrization for temperature and depletion: In order to account correctly for the effects of temperature on diffusion-theory parameters, a feedback model must be included among the methods. Prof. Kostadin Ivanov at the Pennsylvania State University (PSU) has developed and validated a new adaptive high-order table look-up model for cross-section parametrization. PSU will modify their model to be compatible with PEBBED and with the methods developed at Georgia Tech for generating diffusion-theory data.

Development of an automatic optimization routine for efficient and accurate sensitivity studies, design, and fuel management: An automatic optimization routine, based upon a genetic algorithm, will be developed and integrated with PEBBED to perform core design optimization. The algorithm will generate core size and flow parameters for PEBBED input and evaluate user-specified objective functions based upon the resulting PEBBED output.

Exploration of the implications of inhomogeneities in pebble packing: Packing in a cavity filled with identical spheres is not uniform, but varies according to a function that resembles a damped oscillation from zero at the walls to an asymptotic value several sphere diameters away. In small PBRs, where the oscillations may extend over a large fraction of the core diameter, the effects of the oscillations may be significant. Calculations will be performed to assess these effects, and an empirical model will be developed for inclusion in the homogenization codes.

Incorporation of code components into PEBBED: The neutronics analysis of PBRs differs from that of light water reactors because PBR fuel moves and the neutronics and depletion equations are coupled to this motion. The PEBBED code solves this coupled problem. All the new methods and codes developed in the tasks described above will be coupled to or implemented within PEBBED. The resulting code suite will be benchmarked by Prof. Barry Ganapol of the University of Arizona. Verification and validation will be performed using data supplied by the South African company, PBMR (Pty), Ltd., who will be participating as unfunded collaborators.

The team assembled for this project has a unique ability to perform the proposed work. The PIs have developed the PEBBED code and derived the $\theta$-dependent analytical nodal formulation of the diffusion equation. The collaborators at Georgia Tech, with the support of Prof. deOliveira, have a unique and ideally suited approach to homogenization. The collaborators at PSU have a unique and ideally suited approach to data parametrization. Prof. Ganapol is known worldwide as an expert in all kinds of benchmarking problems. PBMR (Pty) Ltd. will provide relevant data from the world's foremost PBR design project.

The project will produce a set of interrelated codes to perform neutronics design and in-core fuel cycle optimization for a PBR quickly and efficiently. In addition, the project will lead to publications for conferences and journals. The new capabilities will be demonstrated on sample optimization problems (PBR fuel cycle and non-proliferation characteristics of discharged pebbles).

## 6. Waste Managment

This program area includes 5 NERI research projects awarded in FY 1999. It addresses the long-term R&D goal related to fuel cycle research, which considers the impact of fuel cycle options on waste generation, waste form, and waste storage and disposal.

Projects currently funded include R&D that addresses nuclear waste with regard to technological improvements in the back-end fuel cycle process. Novel approaches are proposed to reduce the physical volume of spent nuclear fuel and to recycle or reuse spent nuclear fuel without reprocessing in a manner that maintains the highest degree of proliferation-resistance. Additional R&D is being performed in the use of concrete for nuclear waste containment.

# Nuclear Energy Research Initiative

## Directory of Waste Management Project Summaries

NUCLEAR ENERGY RESEARCH INITIATIVE

# Monitoring the Durability Performance of Concrete in Nuclear Waste Containment

Primary Investigator: Franz-Josef Ulm, Massachusetts Institute of Technology

Collaborators: Commissariat a l'Energie Atomique (French Atomic Energy Commission)

Project Number: 99-126

Project Start Date: August 1999

Project End Date: August 2002

## Research Objectives

Concrete is commonly employed in radioactive waste disposal as an effective construction material for containment barriers, liners, and encasement of containers. The objective of this research is to develop the scientific knowledge and the appropriate engineering tools required to evaluate and quantify the durability performance of concrete containment of nuclear waste subjected to the negative chemical-degradation scenario of calcium leaching. Monitoring the durability performance here means the quantitative assessment, in time and space, of the integrity of the container during the entire storage period, and requires the consideration of the multiple couplings between diffusion-dissolution of calcium and deformation and cracking.

With regard to the time-scale, a durable design for waste containers needs to take into account some reference scenario of chemical degradation, in particular the unfavorable one of calcium leaching by pure water. This design scenario refers to the risk of water intrusion in the storage system. For the reference scenario at hand, it is generally assumed that concrete is subject to leaching by a permanently renewed deionized water acting as a solvent. The calcium ion concentration in the interstitial pore solution leads to dissolution of the calcium bound in the skeleton of Portlandite Crystals, $Ca(OH)_2$, and calcium-silica-hydrates (C-S-H), with sharp dissolution fronts. This calcium leaching leads to a degradation of the mechanical properties of concrete (material strength, Young's modulus). Cracks increase the diffusivity of the calcium ions through the structure, and can lead to an acceleration of the chemical degradation, and hence to accelerated aging of the structure. This process can lead to a closed loop of accelerated structural degradation.

## Research Progress

The important scientific findings that will translate into industrial benefits in the field of concrete durability in nuclear waste storage are as follows:

(1) The chemical process of calcium leaching involving kinetics and mineral composition was studied, including the use of alternative leaching agents. State-of-the-art material tests were employed. Based on the scientific analysis of the calcium leaching process, an accelerated material leaching test was conceived and put into practice, allowing for a 300-fold accelerated calcium-leaching process (see Figure 1). This makes it possible to test different kinds of cementitious materials with regard to their leaching characteristics before they are used in industrial applications. Changes in mineral composition due to calcium leaching are now predictable and can be incorporated into industrial planning and design tools. For instance, the thickness of containment structures can be adopted with regard to the demineralization design scenario.

The developed test is now being used in several laboratories internationally for the assessment of the long-term mechanical stability of construction materials.

(2) The role of cracks on demineralization of porous materials was the object of an extensive study. Using dimensional analysis, similar properties that govern calcium leaching and degradation of concrete were thoroughly identified. In particular, studying the similar properties of the governing equations of 'real' calcium leaching in concrete, it was shown that a pure diffusive mass transport through a crack or fracture will not significantly affect the overall degradation kinetics of a

concrete structure. However, it was also shown that advective transport may accelerate the degradation process.

The in-depth analysis of influential parameters concerning cracks in cementitious materials and their consequences on leaching have immediate industrial application. For example, it is now possible to analyze the suitability of a given cementitious material or structure for industrial applications at any moment during its life span. This can improve the decision-making process compared to a decision based solely on the crack size, as is common practice in parts of industry today.

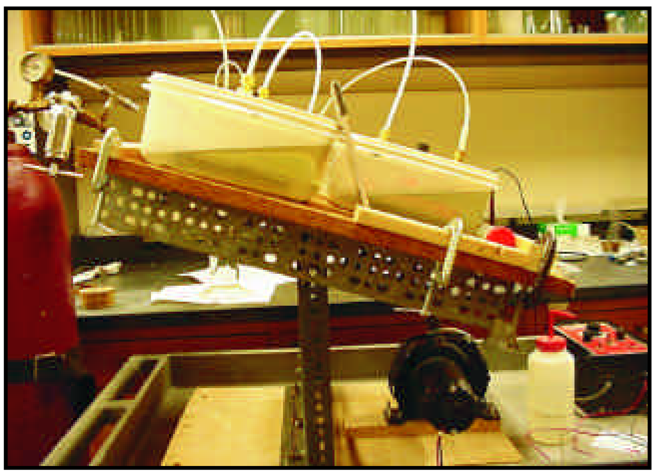
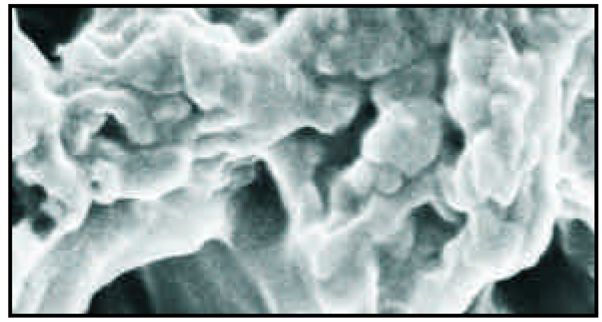


Figure 1. The MIT-NERI accelerated leaching test device uses containers filled with an optimized ammonium- nitrate solution mounted on an oscillating table.

(3) The consequences of calcium leaching on the mechanical behavior of cement paste were identified in mechanical tests, invoking three-dimensional stress states. For the first time, the governing mechanical parameters were identified in 3-D at multiple scales ranging from the nanometer level (see Figure 2) to the centimeter-level of concrete materials. These included the following:

- By means of instrumented nanoindentation, the effect of calcium leaching on stiffness and strength of C-S-H was quantified. It was found that calcium leaching has an important mechanical impact on the so-called outer products of the C-S-H, while the inner products are less affected. This result was a breakthrough in the field of material sciences of cementitious materials (see Figure 3), since for the first time, it was shown conclusively that calcium leaching on the smallest scale accessible to mechanical testing affects only part of the matter. This explains, in part, why calcium-leached, cement-based materials, even when severely leached, still have a residual stiffness and strength.



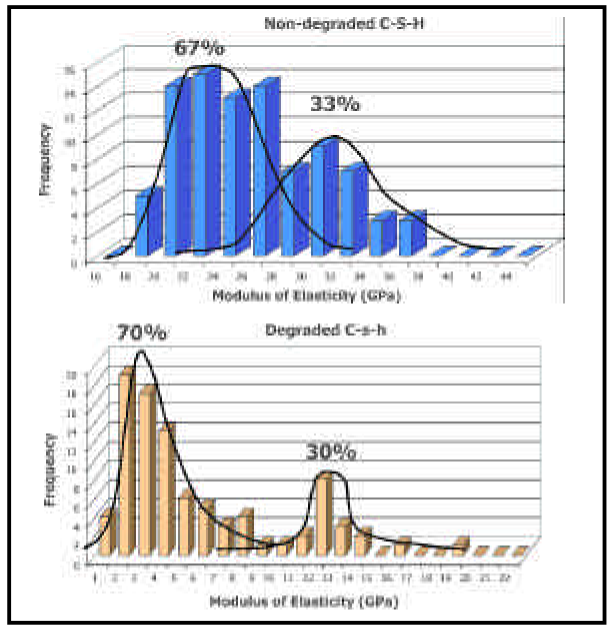



Figure 2. (top) TheSEM-picture is of an artificially aged mortar (asymptotic state), equivalent to 300 to 1,000 years of natural material subjected to "natural" leaching or weathering. Figure 3: (bottom) The graphs present results from 200 nanoindentation tests on non-degraded and degraded C-S-H. The results show that calcium leaching does not affect the volume proportions of the two types of C-S-H, and that the low-density C-S-H (outer products) are heavily affected by calcium leaching, in contrast to the high-density C-S-H.

- By means of macroscopic triaxial compressive tests and direct tensile tests the assessment was completed of the strength and stiffness of asymptotically leached, cement-based materials (cement paste and mortar). This extensive series of tests provided conclusive evidence of the residual-strength domain of calcium-leached materials, which has a relatively low standard deviation, and which therefore can be taken into account in the design of concrete for nuclear waste disposal structures (containment and/or containers).

- While the experimental database was developed for cement paste and mortars, a novel micromechanical model was developed that allows one to upgrade both the stiffness and the strength properties of calcium-leached materials, from the lowest level of cement-based materials to concrete. This novel approach, which was developed for both highly confined stress states and low confinement states, has been validated with the experimental values obtained by nanoindentation and macroscopic material testing. Based on this validation, investigators are confident that the database can now be used for concrete as construction material for containment structures.
- Based on yield design, a safe lower bound solution was developed for the residual design strength of calcium-leached materials, with application in the nuclear waste storage domain. This is a new design concept, and one that should be considered as an economical solution in concrete containment structures for the storage of radioactivity over extended time periods.

(4) The results of mechanical testing are a key element for the mechanical modeling of calcium leaching. This modeling is needed to support industrial monitoring and decision-making regarding safe storage systems. Further aspects of this finding are as follows:

- It is common practice today to assume that because calcium-leached materials are stiff and reduced in strength, they should not be considered mechanically active for the design and operation of concrete employed in nuclear waste storage systems. This will increase the cost of using concrete in nuclear waste storage systems. The experimental and theoretical results of these studies provide clear evidence that asymptotically leached materials have a sustainable residual strength and a stiffness that can be factored into the design and operation of concrete subjected to aging over extended periods of time. This new design concept, once applied, will help to substantially reduce the costs for storage solutions, particularly over longer storage times.
- The multiscale experimental and theoretical approach provides the foundation for optimizing concrete solutions for nuclear waste storage design, starting at the smallest intrinsic level of the material. In particular, given the nanoindentation results, it appears that a concrete with a higher inner-product concentration is much more sustainable, in terms of long-term mechanical performance, than the standard materials employed today.
- The most critical issue in the development of materials and structures for sustainable and economic nuclear waste storage is the predictability of physical states the material may undergo over 300 to 1,000 or more years. The approach developed here, in which time/age is replaced by chemical equilibrium states associated with mechanical performance provides a new basis for the industrial development of such solutions.

(5) The research carried out in the course of this project is now close to delivering a blue-print of the elementary aging components of cement-based materials. These elementary aging components control the mechanical performance of the materials over extended periods of time, although they do not change from one cement-based material to another. Instead, they are intrinsic to the matter, which makes it possible to upscale the micro-behavior assessed by, for example, nanoindentation, to the macroscopic scale, where concrete is employed in nuclear storage systems. This blue-print of the chemomechanical behavior of cement-based materials is the backbone of the micro-chemo-mechanics theory of calcium leaching the researchers have developed for cementitious materials, which ultimately provides the missing link between physical chemistry and mechanics. Further aspects of these novel developments are as follows:

- The engineering implementation of this multi-scale model in a finite-element program makes it possible today to predict and anticipate with a high level of confidence the physical state of concrete materials and structures subjected to calcium leaching over extended periods of time that correspond to the lifetime of nuclear waste storage systems. This finite-element implementation was validated through comparison with experimental structural data, which shows the capacity of the material model to improve the understanding of experimental results at the scales of structures subjected to leaching.

- The model and its implementation form a powerful durability design tool that can be used for the design of new structures and the lifetime analysis of existing nuclear waste storage structures. In particular, by using multi-scale, model-based simulations, it was possible to show that a new generation of ultra-high-performance cementitious materials (UHPC that is currently entering the market offers superior durability performance for nuclear waste storage systems. The sources of the improved leaching resistance of UHPC could be identified and quantified: The absence of Portlandite and the lower porosity significantly slow down the leaching process and reduce the extent of deterioration of the material properties.

(6) The research carried out in the course of this project provides the basis for the development of a new generation of "intelligent" storage systems. In these systems, continuous monitoring of the physical state of the materials and structures, blended with advanced modeling tools developed in this project, will allow the industrial development of the next generation of sustainable and economic nuclear waste storage systems.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Neptunium Speciation in Spent Nuclear Fuel

Primary Investigator: Ken Czerwinski, Massachusetts Institute of Technology

Collaborators: Argonne National Laboratory (ANL), Chemical Technology Division

Project Number: 99-127

Project Start Date: August 1999

Project End Date: August 2002

## Research Objectives

This project will examine the chemical speciation of neptunium in spent nuclear fuel. The R&D fields covered by the project include waste host materials and actinide chemistry. Examination of neptunium is chosen since it was identified as a radionuclide of concern by the NERI workshop. Additionally, information is lacking on the chemical form of neptunium in spent fuel. The identification of the neptunium species in spent fuel would allow a greater scientific-based understanding of its long-term fate and behavior in waste forms.

Research to establish the application and development of X-ray synchrotron radiation (XSR) techniques to determine the structure of aqueous, adsorbed, and solid actinide species of importance to nuclear considerations is being conducted at Argonne National Laboratory (ANL). These studies extend current efforts within the Chemical Technology Division at ANL to investigate actinide speciation with more conventional spectroscopic and solids characterization (e.g. Scanning Electron Microscopy, SEM, Transmission Electon Microscopy, TEM, and X-Ray Diffraction, XRD) methods. This project will utilize all these techniques for determining neptunium speciation in spent fuel.

The chemical species and oxidation state of neptunium will be determined in spent fuel and alteration phases. Different types of spent fuel will be examined. Once characterized, the chemical behavior of the identified neptunium species will be evaluated if this information is not present in the literature. Special attention will be given to the behavior of the neptunium species under typical repository, near-field conditions (elevated temperature, high pH, varying Eh). This will permit a timely inclusion of project results into near-field geochemical models. Additionally, project results and methodologies have applications to neptunium in the environment, or treatment of neptunium-containing waste.

Another important aspect of this project is the close cooperation between a university and a national laboratory. The PI has a transuranic laboratory at the Massachusetts Institute of Technology (MIT) where students can perform spectroscopic and radiochemical experiments. Through the ANL partner, students can have additional experience performing research in a DOE setting. This will provide a unique and constructive opportunity for developing high-quality graduate students with experience and expertise in handling actinides. The ability to produce experienced actinide scientists is currently restricted by the dearth of radiochemistry and nuclear research at universities. Regardless of all else, future researchers must be trained and educated if the United States is to maintain a leadership role in nuclear technology. This project provides such an opportunity.

## Research Progress

Progress on the various tasks of the project will be discussed in turn.

Development of XANES/EXAFS Experiments: The ANL team organized equipment for developing X-Ray Absorbtion Near Edge Structure (XANES)/Extended X-Ray Absorbtion Fine Structure (EXAFS) experiments. The equipment was utilized in the subsequent XANES/EXAFS experiments with Np.

Analysis of Np Solids: ANL prepared and analyzed $NpO_2$. High isotopic purity of Np-237 was used in the preparation of the oxide. The neptunium was reduced to $Np^{4+}$ by taking to dryness in concentrated hydrobromic acid, and the occurrence of complete reduction was verified by absorption spectrometry. The neptunium oxide samples were analyzed by XANES/EXAFS at the MR-CAT[1] beamline to provide reference spectra for future reaction/alteration.

[1] Materials research collaborative access team at the Illinois Institute of Technology.

Phase matching and shifts in the radial distribution of the oxygen in this system will be used to identify and establish the changes in the oxide due to reaction with the air-steam environment.

The ANL team has also preformed theoretical calculations for evaluating XANES/EXAFS spectra. Theoretical calculations on dissolved neptunium species were initiated. The overall goal of this work is to calculate the structures of aqueous neptunium species.

Analysis of Np and Alteration Products: MIT calculated the production of $^{237}Np$ from $^{236}U$. It is intended to produce $^{236}UO_2$ for irradiation in the MIT Reactor. This will produce $^{237}Np$ in the same manner as nuclear fuel. The speciation of Np in the $UO_2$ matrix will be examined by XAFS at ANL. Initial calculations show enough Np can be produced with a two-day irradiation to provide suitable XAFS samples. A sample of $^{236}UO_2$ was created from a $^{236}U$-nitrate solution by precipitation reaction. The precipitate was dried, calcined, and finally sintered under a reducing atmosphere to produce the oxide. X-ray diffraction spectroscopy was performed on a $^{238}UO_2$ standard prepared identically to the $^{236}U$ sample to verify that $UO_2$ was created.

The ANL team examined air-steam reaction on neptunium oxides. The safety review was completed and approved and all vessels needed for the experiments are in hand. The matrix for these experiments was developed, $Np(IV)O_2$ and $Np(V)_2O_5$. The changes in structure and oxidation state in these two solids will be established as a function of temperature (25°C and 150°C), oxygen (air and nitrogen gas) and the presence of water vapor (dry and 100 percent RH). In all cases the changes in structure will be investigated using synchrotron-based techniques (primarily XANES) along with continued characterization of oxide standards.

Examination of Np Chemical Speciation: The sorption of Np to mineral phases was examined as a function of pH, solution, and mineral phase. Goethite, montmorillonite, and tuff were the mineral phases examined. The pH was varied from 5 to 12. Both Ar and air atmospheres were used to analyze the impact of carbonate. The solutions examined were 0.1M $NaClO_4$ and J-13 groundwater. In all case there was sorption of Np to goethite. This sorption was most pronounced under Ar. In the presence of carbonate, in both the air and J-13 conditions, there was less Np sorption to goethite. This indicates the formation of carbonate species, which decrease Np sorption to the goethite. For tuff and montmorillonite, sorption occurred near pH 10 due to Np precipitation. An increase in Np solution concentration was observed above this pH. However, the concentration of Np in solution for all the solids above pH 10 was less than the control solution without solid phases. This indicates the solid phases retain Np at higher pH.

The kinetics of the sorption of pentavalent Neptunium onto Tuff, Montmorillonite, and Goethite mineral phases were investigated in a glovebox under an inert atmosphere by MIT. Two series of samples were run respectively at pH 7 and 9. The ionic strength was set at 0.1M with $NaClO_4$ and the pH was adjusted with 0.1M $HClO_4$ and 0.1M NaOH. Supernatant concentrations were measured by alpha liquid scintillation counting. BET surface analysis provided the specific surface area of Goethite, Tuff, and Montmorillonite, respectively, as 248.06±0.87 $m^2/g$, 163.43±4.63 $m^2/g$, and 193.88±4.50 $m^2/g$. Titration experiments provided the Proton Exchange Capacity to be 0.2275±0.0184 $meq_{OH/gG}$, 0.1645±0.0281 $meq_{OH/gT}$, and 0.2722±0.0675 $meq_{OH/gM}$, respectively for Goethite, Tuff, and Montmorillonite. It was found that the sorption of Neptunium on Montmorillonite was faster than on Goethite, and itself faster than on Tuff. One-term exponential regression fits the kinetics of Montmorillonite while a two-term exponential regression fits those of Goethite and Tuff, assuming that in the last case the surface sorption is fast and occurs within the first 5 hours. Then, a combination of surface sorption and diffusion occurs slower until reaching saturation. In the case of Montmorillonite, a linear regression between the concentration of Neptunium at saturation and the initial concentration of Neptunium has been found to be

$$[Np]_{sat} = 0.0825 * [Np]_{init} + 5 * 10^{-07}; R^2 = 0.9998$$

The calculated distribution coefficients given in $Log(K_d)$ are 3.490±0.210 mL/g for Goethite, 3.4509±0.304 mL/g for Tuff, and 3.477±0.116 mL/g for Montmorillonite. A linear regression shows the relation between PEC and $Log(K_d)$ to be

$$Log(K_d) = -0.2974 * PEC + 3.5578; R^2 = 0.9999$$

The logarithm of complexation constants were evaluated as 6.27, 6.71, and 6.94, respectively, for goethite, tuff, and montmorillonite. Correlations with SSA and PEC were established. The data will be incorporated into geochemical codes.

The ANL team examined the complexation of Np(VI) to organics. It was found that Neptunium (VI), as

$NpO_2^{2+}$, was not stable in the presence of either lactate or acetohydroxamic acid. Slow reduction was observed with lactic acid, presumably due to reaction with the secondary hydroxyl group. For acetohydroxamic acid, reduction to $NpO_2^+$ was instantaneous under all conditions investigated. This fast reduction is typically observed for many amines. Once reduced, Np(V), as $NpO_2^+$, was stable in the presence of both lactic acid and acetohydroxamic acid for periods of weeks. There was no evidence of even trace reduction to form Np(IV) species when lactate was present. Some very slow reduction was noted for acetohydroxamic acid. For this reason, only upper limits for the rate constants were determined. The ANL team also examined the interaction of Np with bacteria.

Performance of Modeling: MIT performed focused speciation calculations on the behavior of Np at Yucca Mountain using the hydro-geochemical equilibrium code CHESS. The influence of the pH on the speciation of Np is studied at different temperatures for a fixed potential. The pH is varied from 4 to 12, the successive temperatures are 25°C, 50°C, 75°C, and 100°C, and the potential is 700mV. The main inorganic ligands taken into account were carbonate and hydroxide. Np(V) and, to a lower degree, Np(IV) complexes dominate the speciation. The temperature and the pH favor the formation of $NpO_2CO_3^-$ and $NpO_2^+$ below pH 10 and $NpO_2OH_{(aq)}$ and $NpO_2(CO_3)_3^{4-}$ above pH 10.

Final activities focused on the XAFS analysis of the Np-containing phase. The ANL team collected spectroscopic data on Np-bearing secondary phases and Np in spent nuclear fuel. The MIT team evaluated the data and determined the Np oxidation state in the material. Additionally, under Task 2, the MIT team incorporated project data into the geochemical code CHESS and performed speciation calculations of Np in the repository near-field.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Experimental Investigation of Burn-up Credit for Safe Transport, Storage, and Disposal of Spent Nuclear Fuel

Primary Investigator: Gary A. Harms, Sandia National Laboratories

Project Number: 99-200

Project Start Date: August 1999

Project End Date: September 2002

## Research Objectives

The Nuclear Energy Research Initiative has funded a critical experiment focused on burn-up credit issues at Sandia National Laboratories. The experiment, when complete, will provide benchmark data that can be used to test the methods and data used in the criticality safety analyses of shipping, storage, and disposal of spent nuclear fuel.

Burn-up credit is the process of accounting for the decrease in the reactivity of spent nuclear fuel produced by the changes in the fuel actinide concentrations and the buildup of fission product absorbers caused by the burning of the fuel. To apply burn-up credit safely, the methods used in its application must be validated: the fuel isotopic composition in the burned state must be accurately predicted, and the neutron multiplication of spent fuel configurations also must be accurately predicted. This project addresses the second part of the validation issue.

A critical assembly with low-enriched $UO_2$ fuel has been built at Sandia. The assembly concept is shown in Figure 1. The figure shows a cut-away view of the assembly core along with a photograph of the assembly. It consists of a water-moderated array of driver fuel elements surrounding a smaller number of experimental fuel elements. Test materials can be inserted between the fuel pellets in the experimental elements to allow accurate measurements of the reactivity worth of important fission products. The assembly also includes three fuel-followed control/safety elements and a fueled source element. Critical experiments can be performed in the unperturbed assembly and with a test material, a fission product simulant, present in the assembly. The benchmark data from the experiment is the difference in the critical array size between the two configurations. The results of criticality safety analysis methods can be compared with the benchmark results to test the analysis methods.

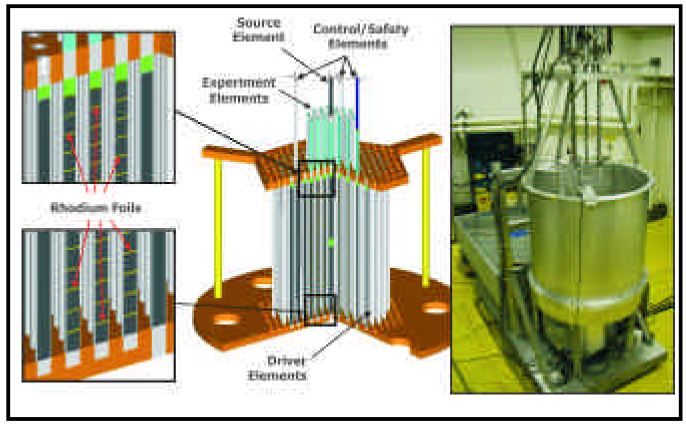



Figure 1. A cut-away view of the Burnup Credit critical assembly is illustrated. The photograph shows the critical assembly tank and associated hardware.

The critical experiment is structured as a three-year project. Five broad tasks were outlined in the project proposal:

- Task 1 - Obtain the necessary National Environmental Policy Act (NEPA) approvals.
- Task 2 - Prepare safety basis documentation for the experiment and obtain approvals.
- Task 3 - Design and procure the experimental hardware.
- Task 4 - Perform the benchmark experiments.
- Task 5 - Decommission and decontaminate the experiment.

## Research Progress

The NEPA approvals for the experiment were obtained in the first year of the project. Sandia filed an Environmental Checklist/Action Description Memorandum with the Department of Energy requesting that the experiment be categorically excluded from further NEPA documentation. The exclusion was granted.

The critical experiments were performed in the reactor room of the Sandia Pulsed Reactor Facility (SPRF) in Sandia's Technical Area V. Early in the project, the determination was made that an addendum to the SPRF Safety Analysis Report (SAR) would be required to establish the safety basis for the critical experiments. The SAR addendum was completed. A document was written that described the Technical Safety Requirements derived from the SAR addendum for the operation of critical assemblies. Both documents were submitted to the Sandia Internal Review and Appraisal System (SIRAS) for safety review. The SIRAS review was completed in April 2001. The SAR addendum and the TSR were then submitted for DOE approval, which was received in December 2001. A three-part readiness assessment was then initiated, which culminated in a review of the facility and the experiment by a DOE team in late February 2002. Issues identified by the DOE readiness assessment team were resolved and approval was obtained to operate the critical assembly.

The design of the critical assembly hardware was completed and procurement of the non-fuel hardware was initiated in the second year of the project. The assembly hardware was received and assembled. Two grid plate sets were obtained, each with different fuel element spacing, to give different neutron spectra in the assembly. The critical assembly is controlled with an instrumentation and control system from an earlier critical experiment that was modified for this project. The driver fuel elements, which were fabricated in 1996 as part of another critical experiment, were transferred from Los Alamos National Laboratory to Sandia. The experimental fuel elements, the control/safety elements, and the source elements utilize the same uranium dioxide fuel stock as that used in the driver fuel fabrication. These fuel elements utilize cladding assemblies fabricated by a commercial nuclear fuel vendor and loaded with fuel pellets at Sandia.

The critical experiments conducted as part of this project were designed to investigate the reactivity effect of rhodium on the assembly. Circular rhodium foils with the same diameter as the fuel pellets were obtained in three thicknesses. The foils are placed between the fuel pellets in the experimental fuel elements. First, the critical array size with only driver fuel was determined. Then the central fuel elements were removed and replaced with experimental fuel elements that had been "poisoned" with rhodium, and the critical array size measured. The benchmark data from the experiment is the difference between the critical array size with the poison and the critical array size without the poison. Experiments with three different foil thicknesses give data for different levels of self-shielding in the foils.

All ten of the critical experiments planned as part of the project have been performed. Five experiments were done with each of the two grid plates: one with driver fuel only; one with experiment elements containing no poison foils; and one with each of the three sets of experiment elements loaded with rhodium foils. During each of the experiments, the critical assembly was loaded in a sequence of steps starting with a low multiplication configuration and proceeding to progressively higher multiplication. Each step provides an updated estimate (by extrapolation) of the number of rods required to achieve a critical configuration. The data from these steps is plotted as 1/M (inverse multiplication) to display the current estimate and guide the determination of the number of rods to load in the next step. Measurements are compared with the results from an extensive series of MCNP calculations to confirm the expected trends. The inverse multiplication graph and a photograph of the critical configuration of the initial BUCCX core, one with only driver fuel, are shown in Figure 2.

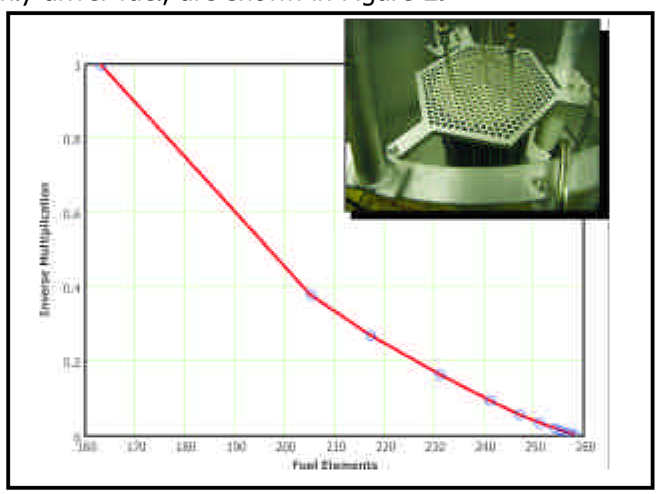



Figure 2. The graph illustrates the inverse multiplication (1/M) plot for the first Burnup Credit critical assembly. The final fuel rod configuration for this core is shown in the photographic insert.

## Planned Activities

The following items will complete the project:

- The final report will document the results of the critical measurements and provide a detailed comparison to the previous analytic results. The critical experiments performed in this project will provide data on rhodium, one of a number of fission products important to full burn-up credit.

Funding will be sought for further experiments using other fission product materials to provide further data in support of efforts to obtain approval for full burn-up credit.

- The experiment will be decontaminated and decommissioned. Since a large fraction of the experimental hardware will be used in another NERI-funded critical experiment, NERI Project 01-124, most of the decontamination and decommissioning activities will be deferred to the completion of subsequent critical experiments.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Deterministic Prediction of Localized Corrosion Damage to Alloy C-22 High Level Nuclear Waste Canisters

Primary Investigator: Palitha Jayaweera, SRI International

Project Number: 99-217

Project Start Date: August 1999

Project End Date: October 2002

## Research Objectives

This research involves developing deterministic models and associated computer codes for predicting the evolution of corrosion damage to high level nuclear waste (HLNW) containers. The principal challenge is to ensure isolation of the waste from the biosphere for periods up to 10,000 years under conditions that can only now be estimated. The lack of relevant databases for the corrosion of candidate alloys means that we cannot rely on empirical methods to provide the design, materials selection, and reliability assessment information. Instead, only strategies based on the employment of deterministic models can be used, because the natural laws (laws of conservation) that are the foundation of these models constrain the solutions to physical reality and are invariant with time.

Existing deterministic models of general and localized corrosion allow us to predict the accumulation of corrosion damage in many systems. However, these models must be customized for predicting damage in HLNW canisters in a tuff repository. Thus, the influence of radiolysis on the corrosion potential and hence on the corrosion rate, for example, must be included in the models. Particular attention must be given to repassivation phenomena, because they eventually determine the extent of damage. Attempts to quantitatively describe localized corrosion damage without proper consideration of repassivation phenomena greatly underestimate the service lives of containers. It is also important to customize the models to the conditions to which the containers are expected to be exposed over their design lives.

The principal objectives of this project follow:

- Develop deterministic models and associated computer codes for predicting the evolution of corrosion damage (i.e., "integrate" damage) to HLNW containers in the Yucca Mountain repository. Corrosion processes that will be considered include general corrosion (oxidation), pitting corrosion, crevice corrosion, and stress corrosion cracking.
- Develop deterministic methods for extrapolating corrosion rate data obtained under "accelerated" laboratory conditions, to the field.
- Use the models to predict the fates of containers after exposure in the repository under various conditions (e.g., humid air, contact with dripping water, repository inundation).

## Research Progress

Damage Function Analysis (DFA), which is based upon the Point Defect Model (PDM) for passivity breakdown, upon deterministic models for cavity growth, and upon the concept of delayed repassivation, has been used to predict damage functions for pitting corrosion in simulated repository environments for times up to 10,000 years. The differential equations for calculating the damage function (DF) have been derived. By analytical or numerical solution of these equations, it is possible to calculate DF under arbitrary conditions if the rate of nucleation and propagation, and the probability of survival of corrosion events are known. The maximum depth of penetration is predicted to be of the order of 1.9 cm, which is of the same order as the design wall thickness of the canister. However, the maximum penetration depth is a sensitive function of the rate constant for the delayed repassivation of active pits, with the maximum depth decreasing as the rate constant increases. No data is currently available for the delayed repassivation rate constant, but methods have been devised in the present program for measuring this important parameter.

A general mathematical model and corresponding computer code have been developed for calculating potential and concentrations distributions in active corrosion cavities. Mass transfer by diffusion and migration, anodic and cathodic processes at the cavity tip and on the sides of the cavity, and hydrolysis and saturation solubility reactions were included in the model. The model also takes into account the potential drop in the external environment (outside the cavity). Based on the general model, a simple, but nevertheless analytical model for calculating potential and concentration distributions, along with cavity propagation rates, has been developed for metals with very low passive corrosion current densities, e.g., Hastelloy C-22. The importance of this development is that it permits simplification of the mathematics, and allows one to predict the potential and concentration distributions without knowing various parameters, such as the equilibrium constants for homogeneous chemical reactions and the kinetic parameters of electrochemical reactions that do not significantly change the concentrations of the principal ionic species in the cavity. The fact that reliable analytical expressions can be obtained for the rate of pit or crevice propagation is very important, because accurate numerical simulation of corrosion damage for a long period (up to 10,000 years) may require a prohibitively large amount of computer time. The conditions that allow the investigator to consider the pit propagation rate as a constant have also been obtained.

Detailed radiochemical simulations of the effects of ionizing $\gamma$ and neutron radiation on the properties and chemical composition of electrolyte solutions under repository conditions indicate that the impact on pH and corrosion potential should be minimal. However, the long exposure times in actual repository systems preclude excluding radiolytic effects completely. Resolution of this issue will require accurate experimental data for the influence of pH and potential on the kinetics of oxygen reduction on Alloy C-22.

It has been shown in this program that, in principle, the possibility that corrosion initiates and propagates on HLNW containers in the Yucca Mountain repository at short times (hundreds of years) when the temperature is significantly above the boiling temperature of water cannot be ignored. This is because the surfaces are covered by highly hydrophilic oxides, hydroxides, and oxyhydroxides that will hydrate to the form of corresponding hydroxides and/or retain water, thereby acting as proton conductors and hence as electrolytes.

Alloy C-22 is found to undergo pitting attack in very aggressive, low-pH, high-chloride, high-temperature environments, with the breakdown potential being near the transpassive dissolution potential. However, because of the distributed nature of the breakdown potential, pits are expected to nucleate at potentials that are significantly more negative than the mean, so that pitting is expected under considerably less aggressive conditions, provided the observation time is sufficiently long. The pits, as observed in short-term experiments, are open and of low aspect ratio (depth/diameter) as shown in Figure 1.

Figure 1. The photomicrograph shows pitting and grain boundary corrosion in Alloy C-22 after exposure to saturated NaCl solution at 80°C under 997 mV (vs. Standard Hydrogen Electode, SHE) applied potential for 10-days (photograph area 75 x 40 μm).

Experimental data is being measured for a variety of model parameters, including the kinetic parameters (exchange current densities and Tafel constants) for the reduction of oxygen and the evolution of hydrogen, and for the oxidation of the substrate, on Alloy C-22 in sodium chloride solution as a function of temperature and pH. The greatest challenge has been to achieve quasi steady-state conditions, to conform to the constraints of the models. Achieving steady-state conditions is particularly difficult with respect to the anodic oxidation current, because of the extraordinarily long time that the transients last. Nevertheless, a reasonably complete set of parameter values is being assembled, which will be of value not only in the present work, but also in studies being carried out elsewhere on the most challenging problem of HLNW disposal.

## Planned Activities

Measurements of key model parameters will be continued during the remainder of the project period. The principal emphasis will be on the measurement of damage functions for Alloy C-22 in well-defined environments under well-defined electrochemical conditions as a function of observation time. The damage functions will allow estimates to be made of a delayed repassivation constant for Alloy C-22 in prototypical repository environments and hence will allow values to be obtained for this most important parameter, as identified by the theoretical work carried out in this program. The value of delayed repassivation constant is very important for predicting the evolution of corrosion damage, because if this value is sufficiently high the probability of survival of a pit over a long observation time is negligible. Considerable effort is also being given to the development the Mixed Potential Model for predicting corrosion potential and general corrosion rate for Alloy C-22. Emphasis in this task is being placed on the measurements of passive anodic current density as function of potential, temperature, and pH (reliable experimental data for cathodic processes on Alloy C-22 has been already obtained in this program). The general computer code for predicting the simultaneous accumulation of general and localized corrosion damage to HLNW canisters will be completed shortly.

NUCLEAR ENERGY RESEARCH INITIATIVE

## A Single Material Approach to Reducing Nuclear Waste Volume

Primary Investigator: James V. Beitz, Argonne National Laboratory

Collaborators: Nuclear Environment Technology Institute (Daejeon, Republic of Korea)

Project Number: 99-219

Project Start Date: October 1999

Project End Date: September 2002

### Research Objective

This project is concerned with developing an innovative single-material, minimum-volume approach for the selective sorption of metal ion radionuclides from aqueous waste solutions and creating a final nuclear waste form that is suitable for long-term storage or geological burial. The project is based on a chemically functionalized porous silica, termed Diphosil. Diphosil was created as an ion exchange medium that selectively and nearly irreversibly sorbs highly charged metal ions, such as actinides, from appreciably acidic aqueous solutions. The chelating power of Diphosil is due to diphosphonic acid groups that are anchored to its silica surface via organic spacer groups. Approximately 90 percent of the weight of dry Diphosil is silica ($SiO_2$).

Underlying this project is the hypothesis that heating metal ion-loaded Diphosil in air will oxidize its organic content to water vapor and carbon dioxide and its phosphonic acid groups to phosphoric acid that would react with the sorbed metal ions to produce metal phosphates. Based on literature reports of the properties of porous silica, it was further hypothesized that additional heating would either volatilize any excess phosphoric acid or cause it to react with the silica to form silicon phosphates. At still higher temperatures, pore collapse should occur, thereby microencapsulating and chemically fixing the sorbed metal ions in phosphate-rich metal phases in vitreous silica. Vitreous silica is one of the most radiation-resistant glasses known.

### Research Progress

Project activities to date have confirmed the hypotheses as to the events that might occur when metal ion-loaded Diphosil is heated in air. The process of converting porous silica to fully dense silica is referred to as thermal densification in the literature because it occurs at temperatures far below the melting point of bulk silica and consequently does not involve a phase change such as melting. The term thermal densification has been adopted to refer to the entire set of processes that occur when metal-ion-loaded Diphosil is heated in air to the point of pore collapse and beyond. The project has investigated the following topics of importance to a single-material approach to reducing nuclear waste volume.

<u>Solution Composition Effect on Metal Ion Sorption:</u> This work has investigated the influence of solution composition variables on sorption of heavy metal ions into a chemically functionalized porous silica (Diphosil). Diphosil has been shown to extract metal ions from aqueous solutions that contain significant concentrations of ethylenediaminetetracetic acid (EDTA) at near-neutral pH. Aqueous solutions of EDTA are frequently used in decontaminating surfaces because of its powerful chelating action for many metal ions. Using laser-induced fluorescence methods, evidence was obtained that Diphosil sorbs trivalent metal ions from concentrated phosphoric acid that contains a small concentration of nitric acid. This mixed-acid media corresponds to the expected composition of the spent working medium of a nitric-phosphoric acid oxidation process for treating organic waste with significant plutonium contamination.

<u>Maximum Metal Ion Loading:</u> To determine the maximum heavy metal ion loading using Diphosil, optical spectroscopy was used to measure metal ion concentration during the sorption process. For example, the maximum uptake of trivalent neodymium ions ($Nd^{3+}$) into Diphosil from dilute nitric acid was determined by monitoring a characteristic near-infrared optical adsorption band of $Nd^{3+}$ and was found to be 2 percent of the dry weight of the Diphosil used.

<u>Densification Optimization:</u> On-line, real-time infrared analysis has been carried out of the gases evolved during

thermal densification of Diphosil in purified air as a function of heating rate and metal ion loading (see Figure 1). Optimal thermal densification conditions have been identified by varying heating and gas flow rates. The resulting material contains the selectively sorbed heavy metal ions in fully encapsulated nanophases that are embedded in nearly colorless, nonporous vitreous silica that is highly resistant to radiation damage. Small-angle neutron scattering studies using silica contrast matching, at the Intense Pulsed Neutron Source at Argonne National Laboratory, provided clear evidence for the nanophase character of metal ion-loaded Diphosil that had undergone thermally densification. Furthermore, the nanophases produced were found to be inaccessible to water and their size dependent on the type of metal ion.

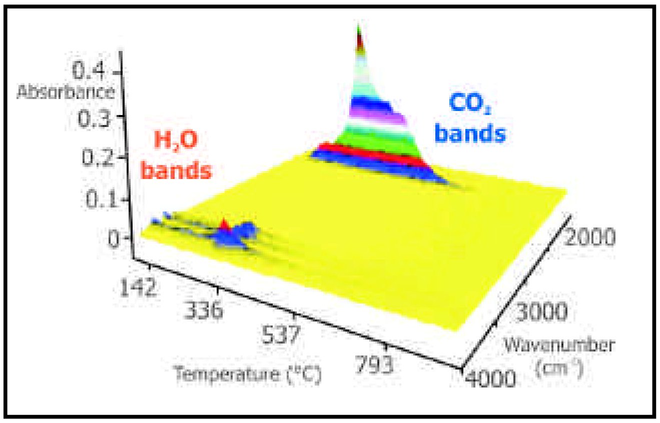



Figure 1. FT-IR absorption spectra of gases evolved as fully uranyl ($UO_2^{2+}$) loaded Diphosil was heated at 2°C/minute in flowing dry, purified air. At this heating rate, the organic content of Diphosil is converted to water vapor and carbon dioxide while its phosphonic acid groups oxidize to phosphoric acid that reacts with $UO_2^{2+}$ and the surrounding silica.

Nonradiative decay: Studies on luminescence decay rates of 3d electron states of a transition metal ion, 4f electron states of trivalent lanthanide ions, 5f electron states of trivalent actinide ions, and charge transfer states of actinyl ions provided rapid means of assessing the proximity of such ions to each other in metal ion-loaded Diphosil prior to and following thermal densification processing. Facile ion-ion energy transfer dominated nonradiative decay processes and resulted in highly nonexponential luminescence decays for each of these cases when Diphosil was loaded with a single type of metal ion, such as $Eu^{3+}$. Studies in which a luminescing metal ion, such as $Eu^{3+}$, and a spectroscopically silent metal ion, such as $La^{3+}$, were simultaneously incorporated into Diphosil, provided a means of controlling ion-ion energy transfer rates and thereby proving that the observed nonexponential decays arose primarily from ion-ion energy transfer. Nonradiative decay studies also provided evidence of complete dehydration of metal ions in thermally densified, heavy metal ion-loaded Diphosil.

Leach rate and radiation damage: Radiation damage studies were carried out on Diphosil into which had been sorbed primarily trivalent lanthanum ions along with lesser amounts of the radioisotopes Cm-245, Bk-249, and Es-253. This material then underwent thermal densification at 1,100°C. Damage resulting from alpha decay of Es-253 (20.5 day half life) to Bk-249 was tracked using time- and wavelength-resolved, laser-induced fluorescence on 5f electron states of $Cm^{3+}$, $Bk^{3+}$, and $Es^{3+}$ ions. These studies showed that decay daughter Bk ions were ejected from the heavy metal phosphate nanophases that contained the parent Es ions and were stopped primarily in the vitreous silica in which those nanophases had been embedded by thermal densification. Calculations of ion stopping distances using TRIM 2000, an ion implantation computer code, support the conclusions we reached from our laser-induced fluorescence studies.

Criticality control: Thermally densified Diphosil contains primarily silicon, oxygen, and phosphorus atoms. As such, it provides some moderation of fission neutrons but will not readily capture thermal neutrons. Anomalous small-angle, X-ray scattering (ASAXS) has been used at the Basic Energy Sciences Synchrotron Radiation Center at the Advanced Photon Source at Argonne National Laboratory to investigate an innovative method for incorporation of gadolinium (Gd) of natural isotopic abundance into Diphosil with no loss of fissile isotope loading. Gadolinium is a nearly ideal criticality control agent for our purposes due to its large thermal neutron absorption cross section. ASAXS provides a means of identifying phases that contain resonant metal ions (Gd in this case) and the size of such phases down to the nanometer scale. ASAXS studies confirmed the expected increase in size of the nanophases that result from thermal densification of Diphosil into which $Gd^{3+}$ ions had been incorporated using our method following full loading of Diphosil with terbium ions bound to diphosphonic acid groups.

Emerging Waste Stream Application: Diphosil possesses the unusual ability to selectively and strongly sorb high valent metal ions, such as actinides, from aqueous solutions. Assessment of the range of aqueous waste solutions that are amenable to processing via Diphosil is being pursued. In particular, spent chloride salts have been identified, such as are generated during electrometallurgical or pyroprocessing of spent nuclear reactor fuel, as an emerging waste stream application for a single material approach to reducing nuclear waste volume. The working medium in present electrometallurgical processing and in many pyroprocesses is a chloride salt or a mixture of chloride salts. Eventually such salts become spent and then must be disposed of although they contain some fission products and actinides.

In the case of such spent process salts, the present high-level reference waste form is glass-bonded sodalite. Unfortunately, glass-bonded sodalite retains the chloride content of the spent salt, which raises concerns about chloride-induced stress corrosion cracking when such a waste form is in contact with stainless steel over prolonged periods of time such as would be the case at the Yucca Mountain geologic repository.

The present work has shown that Diphosil provides sufficient chelating power to remove trivalent lanthanide ions (surrogates for actinide ions such as trivalent americium) from concentrated salt solutions that can be formed by dissolving spent process salts in dilute hydrochloric acid. Rinsing the metal ion-loaded Diphosil with water removed chloride ions and subsequent thermal densification processing chemically fixed and nanoencapsulated the sorbed lanthanide ions in vitreous silica. This project's single material approach to reducing nuclear waste volume is also a promising method for creation of a minimum volume, chloride-free, high-level waste form in the treatment of spent electrometallurgical and pyroprocessing salts.

## Planned Activites

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

## 7. Fundamental Nuclear Science

This element includes 25 NERI projects to date of which 13 were awarded in FY 1999, 1 in FY 2000, 5 in FY 2001, and 6 in FY 2002. It addresses the long-term R&D goal of developing new technologies for nuclear energy applications, of educating young scientists and engineers and training a technical workforce, and of contributing to the broader scientific and technological enterprise.

Today's U.S. reactors, which are based largely on technology from the 1970s, operate under close supervision in a conservative regulatory environment. Although the knowledge base is adequate for these purposes, improvements in the Nation's knowledge base and reduction of the inherent uncertainties concerning nuclear reactors could bring costs savings to current reactor operations and reduce the costs of future reactors. They could also enable innovative designs that reduce the need for excessively conservative and costly safety and reliability factors, and significantly extend safe operating lifetimes. Future reactor technologies are likely to involve higher operating temperatures, advanced fuels, higher fuel burn-up, longer plant lifetimes, better materials for cladding and containment vessels, and alternative coolants. To implement such features, substantial research must be carried out in fundamental science and engineering to supplement applied research on individual promising design concepts. Such fundamental research need not and should not be directed to any specific design. Although motivated in part by the need for new nuclear reactor system designs, the research would also have a far-reaching impact elsewhere in engineering and technology.

The five broad topics identified in the Long-Term R&D Plan related to fundamental nuclear sciences include the following:

- Environmental effects on materials, in particular the effects of the radiation, chemical, and thermal environments, and aging
- Thermal fluids, including multiphase fluid dynamics and fluid structure interactions
- The mechanical behavior of materials, including fracture mechanics, creep, and fatigue
- Advanced material processes and diagnostics
- Reactor physics

Projects currently selected under this element include R&D in fundamental science in the fields of material science, chemical science, computational science, nuclear physics, or other applicable basic research fields. Selected research subjects include irradiation, chemistry, and corrosion effects on nuclear plant materials; advanced new materials research; innovative computational models; and, the investigation of nuclear isomers that could prove beneficial in civilian applications.

NUCLEAR ENERGY RESEARCH INITIATIVE

Directory of Fundamental Nuclear Sciences Project Summaries and Abstracts

NUCLEAR ENERGY RESEARCH INITIATIVE

# Effects of Water Radiolysis in Water Cooled Nuclear Reactors

Primary Investigator: Simon M. Pimblott, University of Notre Dame Radiation Laboratory (NDRL)

Collaborators: Georgia Institute of Technology (GIT)

Project Number: 99-010

Project Start Date: August 1999

Project End Date: May 2003

## Research Objectives

The aim of this project is to develop an experiment- and theory-based model for the radiolysis of non-standard aqueous systems like those that will be encountered in the Advanced Light Water Reactor (ALWR). Three aspects of the radiation chemistry of aqueous systems at elevated temperatures are considered in the project. They are the radiation-induced reaction within the primary track and with additives, the homogeneous production of $H_2O_2$ at high radiation doses, and the heterogeneous reaction of the radiation-induced species escaping the track.

The goals for the latter stages of the program are as follows:

- Development of an algorithm and the testing of code to simulate high-Linear Energy Transform (LET) heavy-ion track structure in water
- Simulation of $H_2$ saturated solutions at ambient temperature
- Development of an experimental protocol for $H_2O_2$ measurement from gamma irradiation
- Measurements of the effect of $H_2$ on $H_2O_2$ yields in gamma irradiated solutions at high doses
- Design of a cell for pulse radiolysis at elevated temperatures
- Irradiation of heavy loaded suspensions of metal oxides in water at ambient temperature
- Determination of the effect of surface potential on escape depth from narrow band-gap oxide materials
- Testing of Electon Parametric Resonance (EPR) and conductivity techniques to measure the charge escape of electrons and holes from these oxides
- Determination of electronic band structures of doped zirconia
- Controlled irradiation of iron oxide covered with water
- Integrity measurements on the zirconia and iron-oxide/water over-layers

## Research Progress

The principal focus of this program is to construct an experiment and theory-based model for the radiolysis of non-standard aqueous systems found in nuclear power plants. The research project has two complementary aspects, one simulation-based and one experimental. A methodology was developed for evaluating the energy loss properties of non-relativistic light and heavy ions in condensed media, and calculations were performed for a variety of radiation types in gaseous and in liquid water. The agreement between calculated stopping powers and ranges and available experimental data is excellent over the range of specific energies (E/amu) of radiation chemical interest, 0.1-100 MeV/amu. The NDRL suite of computer codes, TRACKSIM and TRACKKIN, for the simulation of low-linear energy transfer (LET) track structure and chemistry in condensed media was extended to include radiation particles with moderately high LET in liquid water. These codes address two aspects of the radiolysis: the structure of the radiation track (TRACKSIM) and the chemistry of the resulting spatially non-homogeneous distribution of radiation induced reactants (TRACKKIN). The track structure is simulated using a collision-to-collision methodology, employing the newly developed cross-sections for liquid water mentioned above. The Independent Reaction Times methodology, employed for modeling the radiation chemical kinetics, relies upon the generation of random reaction times from initial coordinate positions from reaction time distribution functions.

To incorporate a significant section of heavy ion track structure, extensive modifications of the simulation methodology were made. Calculations were performed to investigate the radiation chemistry induced by $^{1}H$, $^{4}He$, $^{7}Li$, and $^{12}C$ ions and in aqueous solutions over the temperature range 0°C to 300°C. The results obtained for these well-characterized solutions reproduced yields obtained experimentally (see Figure 1), implying that the technique may be reliably used to model radiation chemical effects in less well-defined, but more relevant systems.

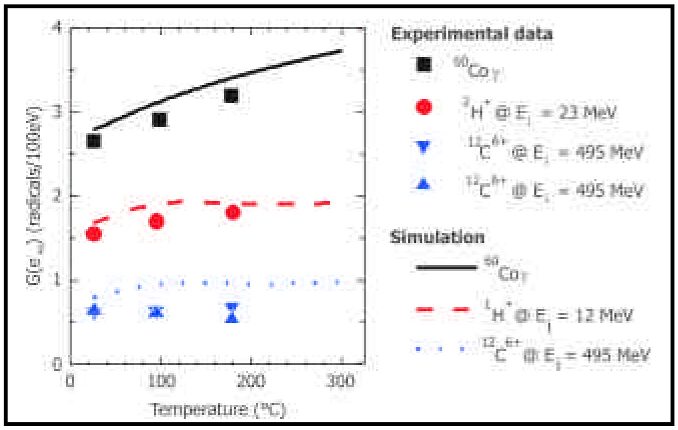



Figure 1. The graph illustrates the effect of temperature on the yield of the hydrated electron produced in the gamma, $^{1}H$ and $^{12}C$ ion radiolysis of water. The points refer to the experimental data of Elliot (1994) while the lines are the predictions of stochastic simulation.

The bulk, homogeneous radiation chemistry of aqueous solutions of hydrated electron and hydroxyl radical scavengers was modeled using yields derived from non-homogeneous stochastic simulations. In some cases, the predictions of these calculations differ from similar more conventional calculations using radical escape yields. These differences are due to intra-track and scavenger chemistry modifying the yields.

One component of the experimental research effort at NDRL has focused on several of the factors that affect the production of hydrogen peroxide in the radiolysis of aqueous solutions. Hydrogen peroxide is the major stable oxidizing product produced in the radiolysis of water and is of considerable interest for the initiation or propagation of corrosion processes. Molecular hydrogen is commonly added to coolant water to lower the steady-state level of hydrogen peroxide. The mechanism for this process was examined with both γ-rays and heavy ions to predict the effects due to the mixed fields associated with reactors. Nuclear reactors typically operate at elevated temperatures so the radiolytic production and decay of hydrogen peroxide was examined up to a temperature of 150°C. Since the radiolytic production of hydrogen peroxide is so important in reactor water chemistry, its mechanism of formation was thoroughly probed using scavengers for both oxidizing and reducing precursors.

The addition of molecular hydrogen was found to significantly lower the steady-state concentration of hydrogen peroxide in the γ-radiolysis of water. This process is due to the initiation of a chain process by the H atoms and OH radicals produced during water radiolysis. Increasing the concentration of hydrogen to its saturation level lowers the hydrogen peroxide concentration to below detectable levels. The results with high LET particles are considerably different. Above about 20 eV/nm the amount of radicals escaping the radiation track is too low to initiate the chain reaction and the steady-state concentration of hydrogen peroxide increases continuously with dose. Simple deterministic models were found to predict the results with γ-rays, but not with heavy ions.

The non-homogeneous nature of energy deposition by ionizing radiation results in high local concentrations of reactive species. An increase in temperature affects both the recombination rates of the reactive species and the diffusive relaxation of the non-homogeneous distribution. In the case of hydrogen peroxide, an increase in temperature decreases its yield due to the lower probability of OH radical combination reactions. The hydrogen peroxide yield at 150°C is only 60 percent of that at 25°C. It was also noticed that hydrogen peroxide is extremely thermal sensitive and readily decays at high temperatures.

In addition to the studies on hydrogen peroxide, pulse radiolysis experiments were conducted on $ZrO_2$ suspensions at high particle concentrations in order to examine the escape of electrons and holes into the aqueous interface. The yield of reducing and oxidizing species was measured in the presence of electron and hole acceptors. At heavy loading of $ZrO_2$, a significant fraction of the energy is absorbed by the solid particles, but the amount of reducing and oxidizing equivalents measured in the aqueous phase and at the interface increases (significantly). This result suggests that energy that was initially deposited in the particles appears in the liquid phase. Furthermore, the increase in the concentration of aqueous reduction and oxidation products is higher than the increase in the energy absorbed by the samples (i.e., higher than the increase in sample density), implying that early-time recombination and trapping processes in $ZrO_2$ particles are less efficient than early recombination in water. Effects of the charge of the scavengers and their interaction with the surface potential were also studied. The yield of electrons escaping the solid is higher than the yield of holes. As a result, charge accumulates in the solid. Contrary to the yield of aqueous redox-radical products, the yield of molecular hydrogen strongly decreases upon increasing $ZrO_2$ concentration. A back-reaction between hydrogen atoms at the interface and the excess holes trapped in the particles is invoked to rationalize this sharp decrease.

The GIT group has completed a series of experiments on proton and molecular desorption from water-covered $ZrO_2$. The results in the high coverage limit are consistent with previously measured yields from amorphous ice. However, the yields in the low coverage limit are very small, indicating that the probability of producing a two-hole state is very low on the $ZrO_2$ surface. This implies that the neutral yields may increase due to single-hole recombination events at the surface, which may resemble an exciton transfer but is actually mediated by a charged carrier. The molecular hydrogen yields were measured as a function of coverage and incident electron/photon energy to examine the importance of exciton, hole, or electron transfer. The proton yields rise monotonically with coverage whereas the HD and $D_2$ yields increase in the manner expected for a two-body or two-step process. In addition, resonance-enhanced multiphoton ionization (REMPI) spectroscopy has been used to measure the yields of the atomic and molecular hydrogen desorbed.

All of the topics mentioned in the program objectives for Phase 2 above have been addressed, although only significant accomplishments have been discussed here. The knowledge obtained is providing a sound basis for Phase 3 of the program.

## Planned Activities

In developing the experiment and theory-based model, two different aspects of the radiation chemistry of water in the nuclear power plant environment will be considered: the initial non-homogeneous reaction of the primary radiation-induced radicals and ions within the radiation track and their reaction with various additives, and the bulk, homogeneous and heterogeneous reaction of the oxidizing and reducing radicals and the molecular products escaping from the track. Thus far, modeling studies have focused on the non-homogeneous and bulk, homogeneous aspects of the radiation chemistry. Final stages of the project will focus on the effects of oxide surfaces and will involve the incorporation into the chemistry model of information about heterogeneous systems that was supplied by the experimental tasks.

Two experimental objectives remain under investigation at NDRL. A remaining goal of the program is the design of a high-temperature radiolysis cell for future studies using high LET radiation. This design is nontrivial due to the thin windows required for heavy ion radiolysis experiments. Experiments will probably be limited to conditions of about 100°C, because of the relatively low ion energies obtained using the local accelerator. In addition, radiation experiments will be performed with surface-modified and core-shell particles, to study specifically the effects of over-layers of silica on hematite and zirconia on the escape of carriers from the particle to the electrolyte, and to determine the effects of these combinations on dissolution rates and on radical rates.

Currently, the GTI group is measuring the yields of the atomic and molecular hydrogen produced during 100 eV electron-beam bombardment on water-covered $ZrO_2$, using REMPI spectroscopy.

Although this research program is scheduled to end in April 2003, numerous avenues for future advances have been opened by the studies performed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Measurements of the Physics Characteristics of Lead Cooled Fast Reactors and Accelerator Driven Systems

Primary Investigator: Phillip J. Finck, Argonne National Laboratory (ANL)

Collaborators: French Atomic Energy Commission, Commissariat à l'Energie Atomique (CEA)

Project Number: 99-039

Project Start Date: August 1999

Project End Date: September 2002

## Research Objectives

Several recent studies in the United States and in other countries have indicated a strong interest in the potential development of lead-cooled critical and sub-critical systems. In order to permit the eventual industrial deployment of these systems, several key technical areas need to be carefully investigated, and solutions for potential technical problems need to be found and implemented.

The neutronic behavior of a lead-cooled fast spectrum system is believed to be relatively poorly known; difficulties arise both from nuclear data uncertainties and from methods-related deficiencies. The French Atomic Energy Commission (CEA) has recognized this situation and has launched an ambitious experimental program aimed at measuring the physics characteristics of lead-cooled critical and sub-critical systems in an experimental facility located at the Cadarache Research Center. A complete analytical program is associated with the experimental program and aims at understanding and resolving potential discrepancies between calculated and measured values. The final objective of the two programs is to reduce the uncertainties in predictive capabilities to a level acceptable for industrial applications.

ANL teams are now participating in the experimental design, measurements, and analytical tasks associated with this effort. In exchange for ANL's participation, all experimental data will be made available to ANL staff.

This program will have three critical outcomes: (1) High-quality experimental data representative of the physics of lead-cooled cores will be available to the U.S. programs, (2) U.S. neutronics codes will be validated for calculating lead-cooled systems, and (3) potential deficiencies in U.S. nuclear data and codes will be identified.

## Research Progress

Efforts this past year were concentrated in two areas: analysis of the MUSE 4 experiment through its associated benchmark configuration, and development of experimental techniques for the measurement of time-dependent data.

Highlights of the progress made follow:

- The first reference configuration of the MUSE 4 experimental program went critical on January 9, 2001. A preliminary analysis of this configuration was performed, followed by the analysis of the international benchmark exercise that was launched in relation with the experimental program (Figure 1 illustrates the configuration used for the study). Deterministic and Monte Carlo methods have been used with JEF2.2, ENDF/B-V and ENDF/B-VI data files. Results obtained by using the cell code ECCO in conjunction with JEF2.2 data are extremely good. Discrepancies have been observed between the VIM Monte Carlo calculations using the ENDF/B-VI data and the corresponding MC$^2$-2 calculations, as well as between the ECCO and MC$^2$-2 calculations with JEF2.2 data. Perturbation calculations have been carried out in order to better understand these discrepancies. They have been attributed to differences in the evaluated data for $Fe^{56}$, $Cr^{52}$, and $Pu^{239}$.

  Following these findings, an attempt was made at first to correct the long-standing deficiency in MC$^2$-2 ultra-fine-group (ufg) scattering cross sections for Fe, Ni, Cr, Mn, and Pb due to their resonance-like structures above the resolved resonance cutoff energies. The correction was necessary to account for the self-shielding effect. Corrections were negligible (less than 100 p.c.m.) for JEF2.2 and ENDF/B-VI data and approximately 200 p.c.m. for ENDF/B-V. Finally the discrepancy was resolved by using the total cross section weighted by the current. This effect is very important for structural material in a reflector medium. When this correction was made, results of $MC^2$-2 agreed with those of ECCO and Monte Carlo codes.

- A major breakthrough was obtained on reaction rate distributions close to the core/reflector boundaries. Up to now, relatively large discrepancies were obtained in the analysis of these quantities. In the course of these studies investigators have shown that using a large number of energy groups (~1,000) significantly improves the agreement between experimental and computational results. The impact of the large number of groups on the eigenvalue calculation is also not negligible for these kinds of configurations where the reflector is in direct contact with the core region and there is no blanket region in the middle. Subsequently, a new computational collapsing procedure has been implemented in order to reproduce the same type of results obtained with a very large number of groups (~1,000), but this time, using a broad-group energy structure (33 groups). The method is iterative and based on the conservation of reaction rates.

Finally, kinetic calculations were performed in order to allow, test, and validate the analysis of the time-dependent experimental results. $^{235}U$ fission rate for several detectors as a function of the time for the deuteron-tritium source pulse in have been calculated. The deuteron beam is assumed to have a time structure of 1 second pulses repeated at 1 kHz. The time step and range were of 1 second and 500 second respectively. Calculations were performed for pulse #1 and for pulse after equilibrium ($n \rightarrow \alpha$). Figure 2 shows time dependent rates using the direct method for the solution of the kinetic neutronic equations. No significant differences were observed when the quasi-static method was used.

Significant spatial effects were observed. The greater the distance of the detectors from the external source (from detectors A, B, C and D in the reflector to the detectors H,J,K and G in the shielding), the more the solution is extended in time. The maximum value is also reached at a different time.

- In the experimental field, the effort has focused on dynamic experimental methods based on the reactor kinetics and neutron noise theory, using time series data. A specific acquisition system has been developed in order to achieve this objective. The time-series-based techniques that have been used are the inverse kinetics method, the pulsed neutron source (PNS) method, the Rossi-$\alpha$ method, and the Feynman-$\alpha$ method. In these dynamic techniques, one makes use of the fact that kinetic behavior in a reactor is related to the reactivity. However, each method is a little different in terms of sensitivity to

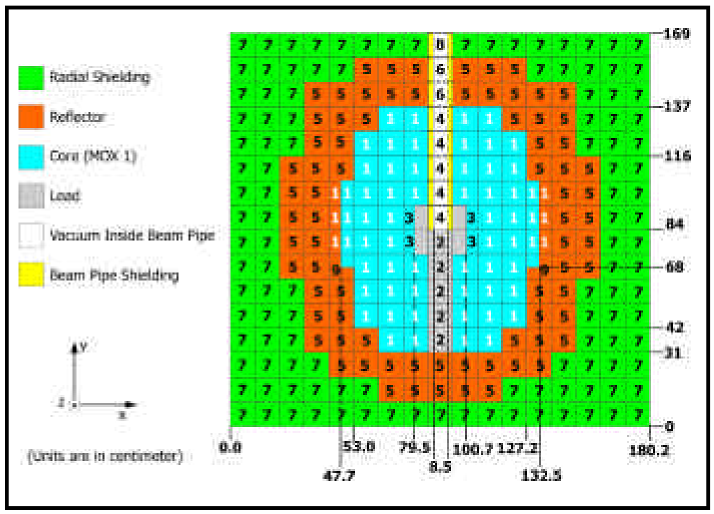



Figure 1: The graphic illustrates the MUSE4 critical (1,112 cells) configuration (top view at half-height)

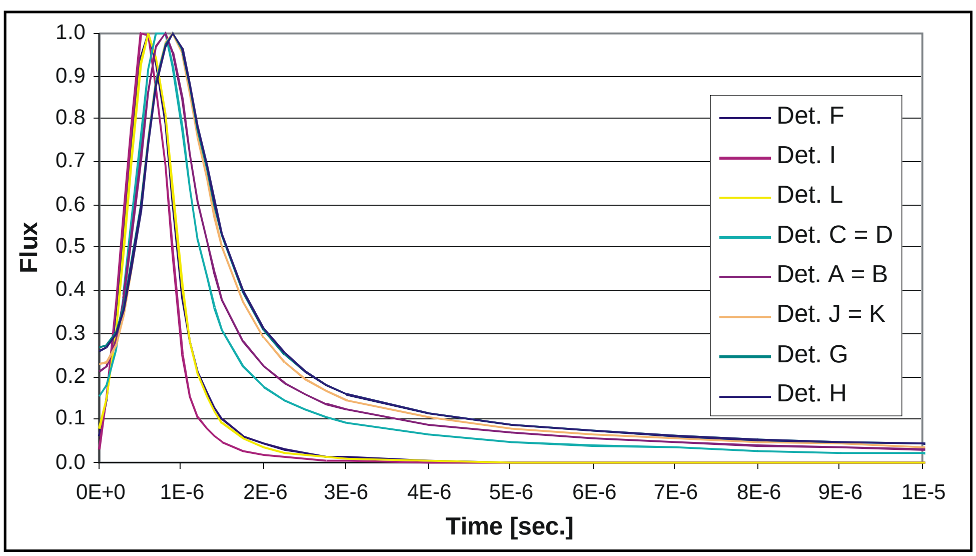



Figure 2. The figure shows time-dependent rates when the direct method is used to solve the kinetic neutronic equations.

other parameters such as background fission rate, detector efficiencies, or kinetic parameters such as the delayed neutron constants.

The PNS, Rossi-$\alpha$, and Feynman-$\alpha$ techniques have been used to infer the ratio $\beta/\Lambda$, a parameter that determines the time scale of kinetic behavior of a reactor system. A fairly large spread (up to 20 percent) has been observed in the determination of this parameter as the system is perturbed by the insertion of a control rod. Two trends were seen in the $\alpha$-values (prompt neutron decay constant). As the reactivity of the system is lowered, lower $\alpha$-values than one would expect are obtained. Additionally, as investigators move from detectors in the core, to detectors in the reactor, and finally to detectors in the shield, a decrease in the $\alpha$-values is also seen. At the present time, the assumption is that this phenomenon is due to the slowing-down of neutrons in the reactor and shield regions, which increases the detector efficiency. This has the effect of increasing the effective generation time in these outer regions.

- The time-dependent measurements have just begun in the MASURCA[1] facility, and the preliminary results are encouraging. However, many more such measurements are needed before investigators can infer the sub-critical reactivity with an uncertainty on the order of 5 percent or less.

## Planned Activities

Although the NERI project has been completed, the ANL activities related to the MUSE experimental program will continue and will be funded under the AFCI program. Both the analysis and experimental tasks will focus on the subsequent subcritical configurations of the program where the GENEPI[2] accelerator will be used in order to insure the sustainability of the system.

---

[1] A fast neutron reactor

[2] A small fast neutron reactor coupled to a deutron accelerator

NUCLEAR ENERGY RESEARCH INITIATIVE

# Mapping Flow Localization Processes in Deformation of Irradiated Reactor Structural Alloys

Primary Investigator: Kenneth Farrell, Oak Ridge National Laboratory (ORNL)

Collaborators: University of Tennessee

Project Number: 99-072

Project Start Date: August 1999

Project End Date: September 2002

## Research Objectives

Ferritic steels, austenitic stainless steels, and zirconium alloys are materials used in the construction of nuclear power reactors and are normally quite ductile and workable. After exposure to neutron irradiation they lose much of their ductility and may even become brittle. This degradation of mechanical properties is governed by the way the metals respond to plastic deformation. In the unirradiated condition they undergo plastic flow homogeneously, and the deformation microstructure consists of uniformly distributed tangled dislocations. After irradiation, the deformation mode is markedly changed to highly localized strain in narrow bands or channels, and sometimes in twin bands. This intensification of strain and stress by dislocation channel deformation (DCD) reduces the work-hardening ability of the metal and causes loss of ductility. The degree of degradation is related to the nature and the details of the dominant deformation mode, which are functions of the radiation exposure and of the mechanical test conditions.

Radiation damage raises the tensile yield strength and ultimate tensile strength (UTS), induces yield point drops in materials that do not normally show sharp yield points, reduces the work-hardening rate and the elongation, and causes premature plastic instability and failure. All of these changes are now known or suspected to involve DCD but only a few quantitative correlations have been made.

Such correlations involve measuring the mechanical properties of the metals as functions of neutron fluence and degree of plastic strain, then performing transmission electron microscopy (TEM) examinations of the strained materials to determine their deformation modes. Maps can then be constructed in which the regions and boundaries of the deformation modes are plotted in terms of plastic strain and neutron fluence. Mechanical properties representing the different deformation modes can be overlaid on the maps, and the maps become pictorial repositories of knowledge relevant to the irradiation behavior of the materials.

The goal of this project is to determine deformation mode maps for A533B ferritic steel, 316 stainless steel, and Zircaloy-4.

## Research Progress

Tensile specimens of the three alloys were irradiated in the hydraulic tube facility of the High Flux Isotope Reactor at Oak Ridge National Laboratory to fast neutron fluences of $6 \times 10^{20}$, $6 \times 10^{21}$, $6 \times 10^{22}$, $6 \times 10^{23}$, and $5.3 \times 10^{24}$ $n.m^{-2}$, E>1MeV, corresponding to nominal doses of 0.0001, 0.001, 0.01, 0.1, and 0.9 displacements per atom (dpa). The irradiation temperature was 65 to 100°C. Post-irradiation tensile properties were measured at room temperature at a strain rate of $10^{-3}$ $s^{-1}$. All three materials underwent progressive irradiation hardening and loss of ductility with increasing dose. Flow stresses were increased, yield point drops were developed, work-hardening rates were reduced, elongations were severely reduced, and early onset of failure occurred by plastic instability. Four modes of deformation identified were three-dimensional dislocation cell formation, planar dislocation activity, DCD (in which radiation damage structure has been swept away), and fine-scale twinning. These modes varied with material, dose, and strain level.

In the body-centered cubic A533B steel, deformation in the unirradiated specimens was homogeneous and occurred by interaction and tangling of dislocations to form dislocation cells (Figure 1). In those specimens of A533B steel irradiated to the two lowest doses, no radiation damage structure (RDS) was detected and there was only minor radiation hardening; the deformation behavior was similar to the unirradiated material. At the middle dose of 0.01 dpa, no RDS was seen but there was considerable radiation strengthening and the work hardening rate was reduced almost to zero. For this dose, the arrangement of strain dislocations was more linear, consistent with the decreased work hardening rate, but there was still some dislocation cell structure. At the highest dose, black spot radiation damage with a mean defect size of 1.3 nm and concentration of about $6.5 \times 10^{22}$ $m^{-3}$ was evident. In the two highest dose specimens prompt plastic instability failures occurred at the yield stress. Some DCD was observed but the strain in the tensile specimens was too highly localized to allow retrieval of truly representative TEM specimens from the deformed regions. The channels were about 40 nm wide.

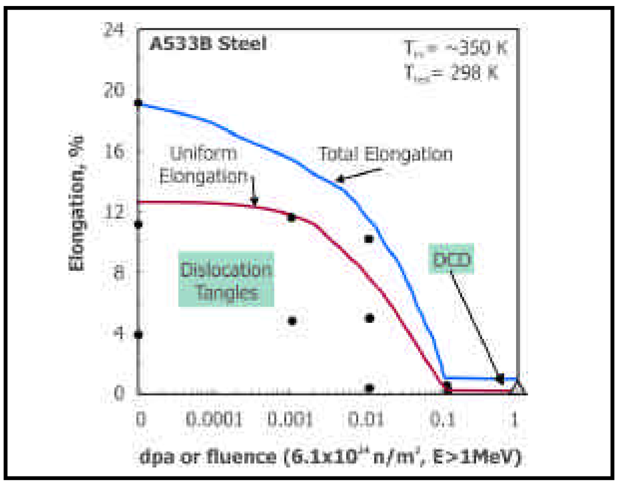



Figure 1. The graph is a deformation mode map for A533B steel neutron-irradiated at 65°C to 100°C and tested at room temperature.

The fluence dependence of the tensile properties of the hexagonal close-packed Zircaloy-4 alloy was found to be of similar form as for the A533B steel, with the exception that the degree of radiation-hardening was higher at the lower doses and lower at the higher doses. No RDS was visible at the lowest dose of 0.0001dpa. At 0.001, 0.01, 0.1, and 0.8 dpa there was fine black spot damage, reaching a size of 1.4 nm and a concentration of $6.1 \times 10^{22}$ $m^{-3}$ at 0.8 dpa. For a dose of 0.001 dpa, and in the unirradiated Zircaloy, plastic deformation during tension testing occurred primarily by coarsely dispersed planar slip in dislocation bands on a single slip system, {1000}<1120> (Figure 2). At 0.01 dpa, the deformation mode was still primarily planar slip, but now occurred on intersecting slip systems, {1000} <1120> and {0111} <1120>. At the two highest doses, where plastic instability failure was entered at the yield stress, the deformation mode was dislocation channeling on the same slip systems. Channel widths were on the order of 50 nm.

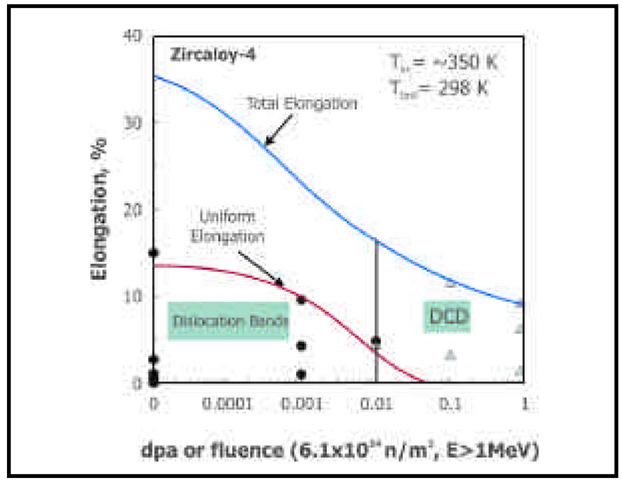



Figure 2. The graph is a deformation mode map for Zircaloy-4 neutron-irradiated at 65°C-100°C and tested at room temperature.

The austenitic 316 stainless steel, which has a face-centered cubic lattice and low stacking fault energy, behaved quite differently from the other two alloys. It displayed a similar degree of radiation hardening as the A533B steel, yet it retained substantial work-hardening and uniform elongation at all doses. In its unirradiated condition, and at the two lowest fluences, where no RDS was visible, it deformed by planar slip on its {111}<110>slip systems (Figure 3). As the level of strain was increased, the slip bands became more pronounced and tangled dislocations appeared in the matrix between the bands. Streaks from fine twins appeared in electron diffraction patterns. Dark field microscopy revealed that the twins were located within the deformation bands. At an irradiation dose of 0.01 dpa, some black spot RDS was found, but the deformation mode was not altered. For the two highest doses, where black spot-type RDS was strong, dislocation channels were cleared through the RDS

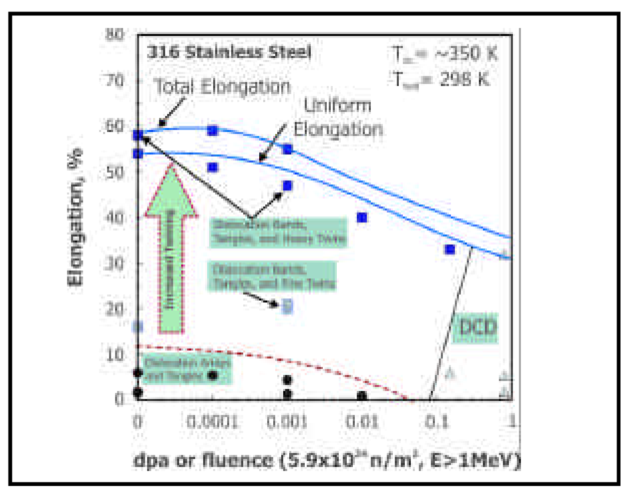



Figure 3. The graph is a deformation mode map for 316 austenitic stainless steel neutron-irradiated at 65°C-100°C and tested at room temperature.

primarily on the easy slip systems. In specimens irradiated to 0.1 dpa, most of the channels were superimposed on the dislocation bands; there were also some very narrow channels containing neither dislocations nor twins. At the highest dose, where the black spot concentration was 4 x $10^{23}$ $m^{-3}$ with a mean size of 1.8 nm, the dislocation bands and microtwins and channels were superimposed in the deformation bands and were very pronounced. Channel widths were about 20 nm. With increasing strain, the blocks of material between the heavy channel bands became subdivided into smaller blocks by development of new channel bands.

The deformation behavior of austenitic stainless steel is different from the other two alloys. Even in its unirradiated condition it deforms in a planar manner. After irradiation, when channels form they are not devoid of dislocations as in the other two alloys. They contain extended dislocations with stacking fault ribbons that are overlapped to form microtwins. Of the three alloys investigated here, the stainless steel has the narrowest channels at a given dose and it is the most resistant to necking. It is suggested that the presence of stacking faults and microtwins in the channels moderates the behavior of glide dislocations in the channels and helps retain some work hardening in the channels, thereby reducing the concentrations of stress and strain in the channels and delaying necking.

Analyses of the fluence ($\phi t$) dependencies of the increases in tensile yield strengths ($\Delta YS$) for all three alloys were made in terms of the relationship $\Delta YS \propto (\phi t)^n$. Values of the radiation hardening exponent, n, were in the range 0.36-0.38 for fluences up to $6 \times 10^{23}$ $nm^{-2}$ (0.1 dpa). Saturation in hardening was noted at higher fluences and was concurrent with acceleration of gross strain localization.

The final phase of the project (Year 3) switched the deformation mapping emphasis to an elevated temperature and higher strain rate. In Years 1 and 2, the irradiations were made at 65°C-100°C and the tensile tests were conducted at a strain rate of $10^{-3} s^{-1}$ at room temperature. For Year 3, the research focused on an irradiation and test temperature of approximately 300°C, and a strain rate of $10^{-1}$ to 1.0 per second. Therefore, new irradiations were made in the HFIR hydraulic tube irradiation facility. A new tensile irradiation capsule was designed to accommodate 14 miniature tensile specimens of each of the three alloys, with a specimen irradiation design temperature of 300°C. The specimens were irradiated to doses of 0.01, 0.1, and 0.8 dpa in March 2002. When the specimens were tensile tested the amount of radiation hardening was considerably less than expected. For the goal irradiation and test temperature of 300°C, published data indicate that a dose of 0.01 dpa should raise the tensile yield strength of annealed stainless steel by about 170 MPa. In the present tests no increase in yield strength was found for the stainless steel and the A533B steel, and only a small increase was discerned in Zircaloy-4. Tests conducted at high strain rates of $100 s^{-1}$ at 288°C, and further tests made at room temperature, showed no significant effects of irradiation on the tensile properties. No radiation damage microstructure and no dislocation channeling were found in TEM of the tested specimens.

Radioactivity readings on the specimens were consistent with their goal neutron exposures. The conclusion is that the temperatures of the specimen in these uninstrumented capsules during irradiation exceeded the goal level and were too high for significant radiation hardening to occur for the investigated damage levels. Postirradiation measurements on passive SiC temperature monitors included in the irradiation capsules indicate that the specimen temperatures were probably in the range 350°C-400°C. This is consistent with sparse literature results showing that the temperatures at which self-annealing of radiation damage will occur during irradiation are 350°C-400°C for the three alloys. Investigation of the cause of overheating during irradiation has focused on two potential culprits: an error in the thermal/hydraulics calculations used to design the internal configuration of the capsule, and unusually low readings of coolant water flow noted in the hydraulic tube system since the reactor cycle in which the capsules were irradiated. Reanalysis of the capsule thermal/hydraulics calculations indicates that the original design had sufficient leeway to allow the specimen irradiation temperature to reach 20°C-30°C higher than desired. Thermal/hydraulics calculations for the low flow condition indicate that the specimen temperature could have increased by approximately 60°C compared to the standard flow condition. The question of reduced coolant flow rate will not be resolved until January 2003 when new flow gauges will be installed in the hydraulic tube irradiation facility. Although these Year 3 results are disappointing for the program goals, they do provide valuable information for irradiation temperature and dose regimes where very little data currently exist; they demonstrate that the upper temperature limit of radiation damage and radiation hardening in these materials at modest neutron doses is only about 350°C.

## Planned Activities

The NERI Project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

## A Novel Approach to Materials Development for Advanced Reactor Systems

Primary Investigator: Gary S. Was, University of Michigan

Collaborators: Pacific Northwest National Laboratory, Oak Ridge National Laboratory

Project Number: 99-101

Project Start Date: September 1999

Project End Date: August 2002

### Research Objectives

Component degradation by irradiation is a primary concern in current reactor systems as well as in future reactors with advanced designs and concepts where the demand for higher efficiency and performance will be considerably greater. In advanced reactor systems, core components will be expected to operate under increasingly hostile conditions (temperature, pressure, radiation flux, dose, and other parameters). The current strategy for assessing radiation effects in order to develop new reactor materials is impractical because the costs and time required to conduct reactor irradiations are becoming increasingly prohibitive, and the facilities for conducting these irradiations are becoming increasingly scarce. Although the next-generation reactors will be designed for more extreme conditions, the capability for assessing materials is significantly weaker than it was 20 years ago. Short of building new test reactors, advanced tools and capabilities are needed now for studying radiation damage in materials that can keep pace with design development requirements.

The most successful of these irradiation tools has been high-energy (several MeV) proton irradiation. Proton irradiation to several tens of displacements per atom (dpa) can be conducted in a short amount of time (weeks), with relatively inexpensive accelerators, and result in negligible residual radioactivity. Together, these factors provide a radiation damage assessment tool that reduces the time and cost to develop and gauge reactor materials by factors of 10 to 100. What remains to be accomplished is the application of this tool to specific materials problems and the extension of the technique to a wider range of problems in preparation for developing and assessing advanced reactor materials.

The objective of this project is to identify the material changes following irradiation that contribute to stress corrosion cracking (SCC) of stainless steels, embrittlement of pressure vessel steels, and physical and mechanical property changes of Zircaloy fuel cladding. Until such changes are identified, no further progress can be made on developing mitigation strategies for existing core components and radiation-resistant alloys or microstructures that are essential for the success of advanced reactor designs.

### Research Progress

Progress is reported separately for the three materials under study.

Stainless Steels: A set of five hardened conditions of commercial 304SS was studied in which the level of hardening remained fixed while the contributions from irradiation and cold work varied. As illustrated in Figure 1, combinations of cold work and proton irradiation were used to achieve a fixed hardness increase of about 180 kg/mm$^2$ for a total hardness of 380 kg/mm$^2$. Proton irradiation was conducted with 3.2 MeV protons at 360°C at a rate of 7 x $10^{-6}$ dpa/s on samples that were previously cold-worked. Magnetic susceptibility tests verified the absence of martensite following cold work. The specimens were then subjected to stress corrosion cracking (SCC) tests in 288°C water, typical of normal water chemistry (NWC) in boiling water reactor (BWR) service conditions. Only the 0 percent cold work + 1.67 dpa and 10 percent cold work + 0.55 dpa samples exhibited irradiation assisted stress corrosion cracking (IASCC), despite the fact that all samples were at a near constant hardness of 380 kg/mm$^2$ ($\pm$5 percent). All other samples failed without any evidence of intergranular (IG) cracking. This result suggests that radiation hardening, in contrast to cold working, is most important in the IASCC process.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Complete Numerical Simulation of Subcooled Flow Boiling in the Presence of Thermal and Chemical Interactions

Primary Investigator: Vijay K. Dhir, University of California, Los Angeles

Project Number: 99-134

Project Start Date: August 1999

Project End Date: April 2003

## Research Objective

The key objective of the proposed research is to develop a mechanistic basis for the thermal and chemical interactions that occur during subcooled boiling in the reactor core. The axial offset anomalies (AOA) are influenced by local heat flux for subcooled nucleate boiling, the nucleation site density on the fuel cladding, and the concentration of boron and lithium in the primary coolant. The approach proposed in this work is very different from that employed in the past, in that complete numerical simulations of the boiling process are to be carried out along with thermal, hydraulic, and concentration fields in the vicinity of the cladding surface. This approach is considered to be the only viable one that can provide, simultaneously, a mechanistic basis for the portioning of the wall heat flux among vapor and liquid and the concentration of boron and lithium, at, and adjacent to, the heated surface. The model is to be validated with data from detailed experiments.

A building block type of approach will be used. By starting with a bubble at a single nucleation site, the complexity of the numerical model and experiments will be increased to include merger of bubbles at the wall as well as interaction of the detached bubbles with the bubbles present on the heated surface. The concentration of boron and system pressure will be important variables of the problem.

## Research Progress

Since the start of the project, both the numerical and experimental effort has been expanded to develop a mechanistic basis for thermal and chemical interactions that occur during subcooled boiling in the core of a nuclear reactor. The numerical effort has so far been focused on a single vapor bubble in either pool or flow boiling. In carrying out the numerical simulations, the conservation equations of mass, momentum, and energy for the two phases along with the conservation of species equation for chemicals present in water have been solved simultaneously. The level set method is used in the numerical simulation. The results of numerical simulation using orthoboric acid as the chemical species present in water reveal that during growth and departure phases of a bubble, the concentration of orthoboric acid varies both spatially and temporally. The highest concentration occurs adjacent to the vapor liquid interface. In regions very close to the wall, this concentration can exceed the solubility limit. In flow boiling, the bubbles are found to slide along the surface before lift-off. The bubble growth continues from departure of the bubble from the cavity to lift-off.

An experimental apparatus for the flow boiling studies has been developed and experiments using silicon strips made from a polished silicon wafer with a microfabricated cavity have been performed. The strips are heated with strain gage heaters that are bonded on the back side. To measure the transient concentration of boron during the bubble evolution, a special probe was developed. Figure 1 shows the measured concentration as a function of distance from the interface when the liquid pool had a boron concentration of 3,000 ppm. Consistent with numerical predictions, the highest concentration occurs near the vapor-liquid interface and decreases with increased distance from the interface. However, the highest concentration observed experimentally near the interface is smaller than that predicted from numerical simulation. One reason for the difference is that the probe yields a concentration that is averaged over a larger volume than calculated through the computations. Plate-out of boron on the silicon surface in the vicinity of a single nucleation site after three hours of subcooled boiling is shown in Figure 2. The plate-out is off-center of the cavity because of the asymmetric growth of the bubble under flow boiling conditions. Most of the plate-out occurs because of evaporation in the microlayer underneath the bubble.

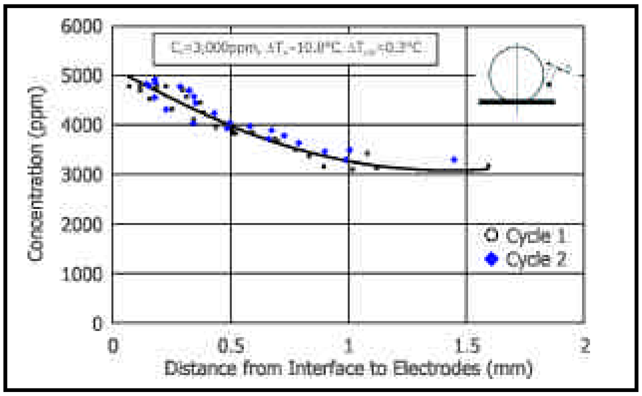



Figure 1. The graph illustrates the measured concentration vs. distance from interface to electrodes.

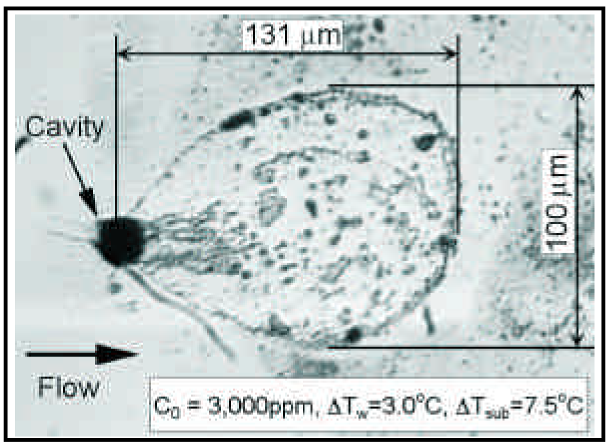



Figure 2. The photo shows boron deposition in subcooled boiling.

Experiments with boron in water were also carried out on a 9 rod bundle. It was found that with 7,000 ppm of boron in the solution, boron crud on the zircalloy cladding was found to reach a thickness of about 30 μm, after 12 hours of boiling on the surface. Because of increased nucleation site density with boron deposits on the cladding, nucleate boiling heat transfer was higher than that on a clean surface. The single phase heat coefficient, however, was lower because of additional thermal resistance of the crud.

A paper based on numerical work was presented at the 2001 IMECE conference; another paper documenting experimental work will be presented at the 2002 IMECE.

## Planned Activities

During the remaining period of the project life, effort will be expanded to carry out numerical simulations of flow boiling with boron. The numerical simulations will be carried out for a range of subcoolings in which bubbles either remain attached to the surface or slide and lift off from the surface. For both cases, the possible plate-out of boron on the solid surface will be quantified. The experimental effort will be expanded to determine the thickness of the boron crust on the test surface and a comparison will be made of the predictions from numerical simulations with the data.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Developing Improved Reactor Structural Materials Using Proton Irradiation as a Rapid Analysis Tool

Primary Investigator: Todd R. Allen, Argonne National Laboratory-West

Collaborators: University of Michigan

Project Number: 99-155

Project Start Date: August 1999

Project End Date: March 2003

## Research Objective

The overall goal of the project is to develop austenitic stainless steel structural materials with enhanced radiation resistance. For this project, the term "radiation resistance" is being used to describe resistance to dimensional changes caused by void swelling and resistance to material failures caused by irradiation-assisted stress corrosion cracking (IASCC). IASCC has been linked to both hardening and changes in grain boundary composition during irradiation. To achieve such enhanced radiation resistance, three experimental paths have been chosen: bulk composition engineering, grain boundary composition engineering, and grain boundary structural engineering. The program involves the use of high-energy proton irradiation as a rapid screening tool to systematically test combinations of alloy composition and thermomechanical treatment conditions to isolate the controlling mechanisms and develop an understanding of how these factors can be engineered to improve material properties.

The alloys chosen for the study have been modeled after commercially available grades of stainless steel commonly used in reactor applications. The model alloys include the following nominal compositions: Fe-18Cr-8Ni-1.75Mn (base 304), Fe-18Cr-40Ni-1.25Mn (Base 330), Fe-18Cr-9.5Ni-1.25Mn + Zr additions (Base 304 + Zr), Fe-16Cr-13Ni-1.25Mn (Base 316), Fe-16Cr-13Ni-1.25Mn +Mo (Base 316 + Mo), Fe-16Cr-13Ni-1.25Mn +Mo + P (Base 316 + Mo + P). Each of the alloying additions was chosen for a specific purpose. Fe-18Cr-40Ni-1.25Mn was chosen because higher bulk nickel concentration is known to reduce swelling, but its effect on IASCC is unknown. Fe-18Cr-8Ni-1.25Mn+Zr alloys were chosen because Zr is an oversized element that might trap point defects and prevent swelling, grain boundary segregation, and other radiation damage. Fe-16Cr-13Ni-1.25Mn, Fe-16Cr-13Ni-1.25Mn+Mo, and Fe-16Cr-13Ni-1.25Mn+Mo+P were chosen to determine why 316 stainless steel is more resistant to swelling and IASCC than 304 stainless steel. The alloys are naturally classified in three groups: the "316 series" (Fe-16Cr-13Ni-1.25Mn, Fe-16Cr-13Ni-1.25Mn+Mo, and Fe-16Cr-13Ni-1.25Mn+Mo+P), the "Zr series" (Fe-18Cr-8Ni-1.75Mn and Fe-18Cr-8Ni-1.75Mn+Zr), and the "Ni-series" (Fe-18Cr-8Ni-1.75Mn, Fe-16Cr-13Ni-1.25Mn, and Fe-18Cr-40Ni-1.25Mn).

## Research Progress

In the first year of the project, the bulk composition engineering path was emphasized. Fe-18Cr-8Ni-1.25Mn, Fe-18Cr-40Ni-1.25Mn, Fe-18Cr-8Ni-1.25Mn+Zr, and Fe-16Cr-13Ni-1.25Mn were studied to determine the effect of bulk composition on swelling and radiation-induced segregation (RIS) at grain boundaries. Samples were irradiated using 3.2 MeV protons at 400°C to 1 displacement per atom (dpa). Swelling was characterized by measuring the void size distribution using a transmission electron microscope (TEM). Radiation-induced grain boundary segregation was measured using a field emission gun scanning transmission electron microscope (FEG-STEM). Microhardness measurements were performed on irradiated and non-irradiated alloys to estimate the effect of irradiation on strength.

Results revealed that alloys with greater bulk nickel concentration have greater RIS. They also have increased hardening and Cr depletion, theoretically making the alloy more susceptible to IASCC. Molybdenum additions did not have a significant impact on the swelling and RIS behavior of the 316 series model alloys, but the addition of phosphorus led to a substantial refinement of the dislocation microstructure, suppression of void formation, and a reduction in the extent of Cr depletion at grain boundaries.

During the second year of the project, the effect of pre-irradiation heat treatments on thermal non-equilibrium grain boundary segregation and subsequent radiation-induced grain boundary segregation in the 316 series of model austenitic stainless steels was studied as part of the grain boundary composition engineering path. The alloys were heat-treated at temperatures ranging from 1,100°C to 1,300°C and quenched using four different cooling paths (furnace cool, air cool, water quench, and ice brine quench) to evaluate the effect of annealing temperature and cooling rate on pre-irradiation grain boundary chemistry. Subsequent RIS behavior following irradiation with high-energy protons was characterized to understand the influence of alloying additions and pre-irradiation grain boundary chemistry in irradiation-induced elemental enrichment and depletion profiles. The study reveals that faster cooling rates provided by water and salt-brine

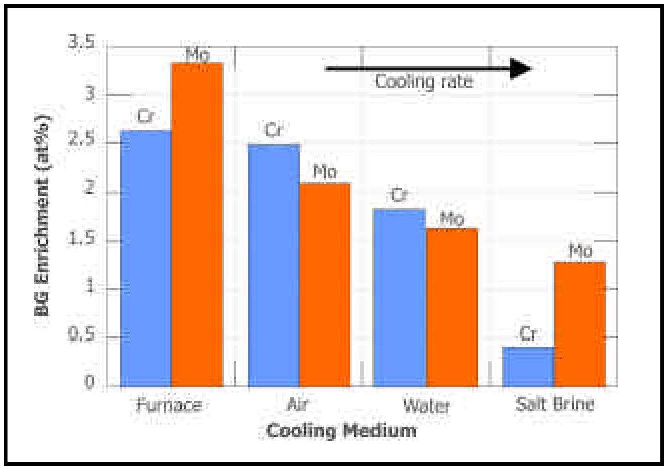



Figure 1. The figure shows the effect of cooling rate on the extent of grain boundary elemental enrichment in Fe-16Cr-13Ni + Mo + P annealed at 1,200°C for 1 hour.

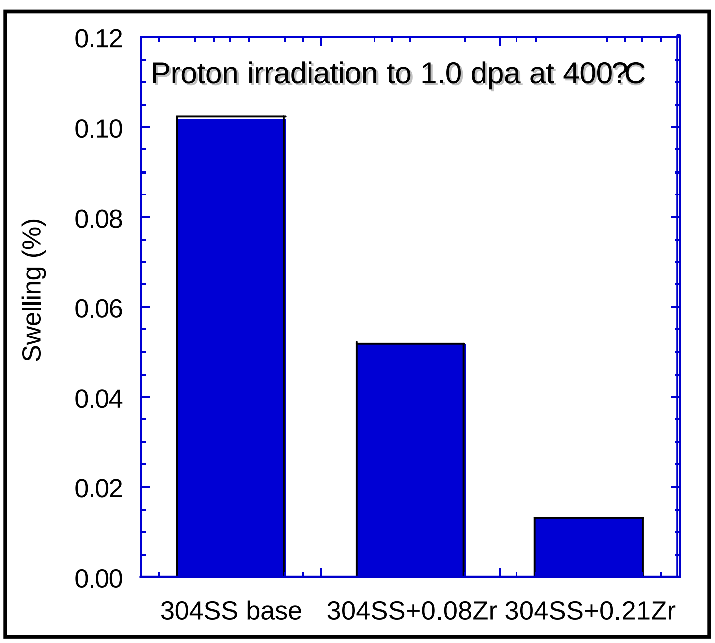



Figure 2. Increasing the Zr content in 304 stainless steel decreases the amount of void swelling

quenching resulted in moderate Cr enrichment. However, slower cooling rates provided by both air and furnace cooling led to more substantial grain boundary enrichment of Cr, and Mo, and depletion of Ni and Fe.

Figure 1 illustrates the change in Cr and Mo enrichment as a function of cooling rate. Lower annealing temperatures also tended to enhance the degree of boundary enrichment. Subsequent proton irradiation of the Fe-16Cr-13Ni + Mo alloy following heat treatments to enrich the grain boundary resulted in the formation of a W-shaped Cr segregation profile and a reduction in the extent of Cr depletion.

Formation of vacancy concentrations during higher temperature annealing and their subsequent migration to sinks during cooling is believed to be the primary process leading to the enrichment of solute species at grain boundaries. Grain boundaries act as sinks for vacancies during cooldown, and the vacancies can drag the solute and thus enrich the boundaries. Models that have been used to describe and explain this include terms for the diffusion of vacancies and vacancy solute complexes to the grain boundary and an associated back-diffusion of free solutes that limits the overall amount of segregation. However, current models do not adequately predict the subsequent segregation behavior during irradiation and the result of this study may provide additional insight into these processes.

In the third year, the radiation response in the 304 + Zr alloys and the 316 + grain boundary composition engineering samples were analyzed.

Following proton radiation, hardness was measured and microstructures were characterized. The addition of Zr decreased the hardening, reduced the swelling (figure 2), reduced the density of radiation-induced void and dislocation loops, and increased the radiation-induced grain boundary segregation. The Zr additions appear to be greater improvement in radiation resistance than the increases in nickel concentration pursued in year 1. Both treatments reduced the swelling, but the Zr-doped alloys did so with out an associated increase in hardening. The 316 plus grain boundary composition engineering delayed the Cr depletion that occurs under irradiation as compared to the non-treated samples. This treatment alone does not provide radiation resistance but may be combined with other treatments to create a more optimal situation.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# An Investigation of the Mechanism of IGA/SCC of Alloy 600 in Corrosion-Accelerating Heated Crevice Environments

Primary Investigator: Jesse B. Lumsden, Rockwell Science Center

Project Number: 99-202

Project Start Date: August 1999

Project End Date: March 2003

## Research Objectives

Most corrosion damage in nuclear steam generators has occurred in tubes forming the tube/tube support plate (T/TSP) crevices. Tubing in this location has experienced damage by pitting, wastage, and intergranular attack/stress corrosion cracking (IGA/SCC), caused by secondary water impurities that have concentrated in these deposit-filled crevices by a thermo-hydraulic mechanism. Crevice chemistries in an operating steam generator cannot be measured directly because of their inaccessibility. In practice, computer codes, which are based upon hypothesized chemical reactions and thermal hydraulic mechanisms, are used to predict crevice chemistry. In most cases the codes have not been benchmarked. Remedial measures for IGA/SCC in the form of water chemistry guidelines have been implemented aimed at controlling crevice chemistries. The guidelines have been formulated based on SCC tests using static autoclaves containing solutions, assumed to duplicate those found in crevices.

The objective of the Rockwell program is to provide an experimental base to benchmark crevice chemistry models, to benchmark crevice chemistry control measures designed to mitigate IGA/SCC, and to model IGA/SCC processes. The objective includes identifying important variables, including the relationship between bulk water chemistry and corrosion accelerating chemistries in a crevice. One important result will be the identification of water chemistry control measures needed to mitigate secondary side IGA/SCC in steam generator tubes. A second result will be a system, operating as a side-arm boiler, which can be used to monitor nuclear steam generator crevice chemistries and crevice chemistry conditions causing IGA/SCC.

## Research Progress

The key element in the approach is a heated crevice apparatus constructed under the NERI program. This is an instrumented replica of a steam generator tube/TSP crevice and operates at simulated steam generator thermal conditions. The pressure in the autoclave containing the crevice is adjusted to give a boiling point of 280°C for the constantly refreshed feedwater. A cartridge heater inside the tube supplies a 40°C superheat, simulating the thermal conditions of the primary water. The apparatus is instrumented to monitor the electrochemical potential (ECP) of the free span, the ECP in the crevice, and the temperature in the tube wall at different elevations in the crevice. The tube is pressurized with He to provide a hoop stress. The additional capability to measure electrochemical noise (EN) monitors IGA/SCC and other corrosion processes on the tube. A schematic of the instrumented crevice and the steam generator tube/TSP replicated is shown in Figure 1. The ZRA is a zero resistance ammeter, which measures the direct current and current fluctuations between the tube and the TSP.

Several advances have been made since the initiation of this project. Using feedwater with sodium hydroxide, work in this program established that the corrosion damage to the tube in the heated crevice duplicates that observed in tubes forming the tube/TSP crevice in operating nuclear steam generators, believed to have caustic crevice solutions. This suggests that the same mechanism causes caustic IGA/SCC in steam generators and in the accelerated conditions of the heated crevice. It was also demonstrated that the relationships between ECP, crevice chemistry pH, and SCC susceptibility follow the thermodynamic/passive film stability methodology. This is of importance because computer codes for crevice chemistry and predictive models rest on the assumption that equilibrium thermodynamics can be applied. The results in this program have established that EN detects the initiation of SCC and monitors crack propagation in Alloy 600. Finally, all results are consistent with the oxide film rupture/anodic dissolution model for IGA/SCC. An understanding of the mechanism of IGA/SCC is necessary for confidence in the success of measures implemented to control and mitigate IGA/SCC.

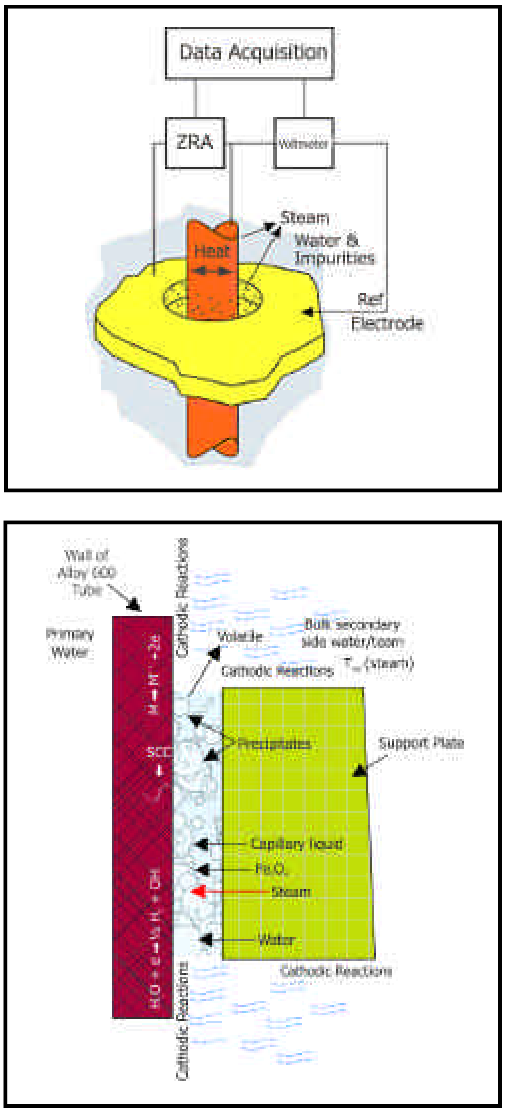



Figure 1. The schematic illustrates (top) the instrumented heated crevice and (bottom) a tube/TSP crevice in an operating pressurized water reactor (PWR) steam generator.

High alkalinity in caustic tube/TSP crevice chemistries was one of the first causes of IGA/SCC in Alloy 600 steam generator tubes in modern operating plants. Accordingly, initial work with the heated crevice investigated caustic IGA/SCC using deaerated feedwater with 40 ppm NaOH. The crevice was packed (22 percent porosity) with magnetite powder, simulating the magnetite deposits and precipitates in the tube/TSP crevices of operating plants. Solution extraction showed that a steady-state concentration of approximately 30 percent NaOH was reached in the crevice after the tube heater was on for approximately 20 hours. These test conditions produced multiple axial cracks, one of which propagated through the wall in four to eight weeks. A destructive examination showed that the cracks were totally intergranular. The crack distribution and morphology are like that found in tubes removed from operating steam generators. An Auger electron spectroscopy analysis of areas on the fracture face showed that the surface was rich in Ni, which is in accordance with thermodynamic predictions for high pH environments. Both Ni or NiO are thermodynamically stable at high pHs, while only soluble species of the alloying metals, Cr and Fe, are thermodynamically stable at caustic conditions. Conditions of high pH lead to the selective dissolution of Cr and Fe. The same surface composition has been found on fracture faces in tubes, forming tube/TSP crevices, removed from operating steam generators believed to have had caustic crevices.

Computer codes describing crevice chemistry and models for IGA/SCC in Alloy 600 steam generator tubes are based on the assumption that equilibrium thermodynamics can be applied. One of these codes, MULTEQ, was used to determine the pH in the heated crevice using the chemistry of the extracted solution. An important result was that the pH determined by MULTEQ agreed with a calculation of the pH derived from the crevice ECP, using an expression derived from equilibrium thermodynamics. Since the results from the two approaches were equivalent, it suggests that the assumptions used in the computer code apply correctly to the conditions in steam generator crevices. Thermodynamics has been used to form a framework for a predictive model for IGA/SCC susceptibility of Alloy 600. This model hypothesizes that the film rupture/anodic dissolution mechanism is responsible for caustic cracking and that IGA/SCC occurs in a zone of pHs and ECPs. The measured value of the crevice ECP when SCC occurred in the heated crevice was well within this zone, supporting this thermodynamic-film instability model. Aggressive impurities in the magnetite deposits and feedwater shifted the "cracking zone" to lower potential values.

The EN, the random pulses of current and ECP generated during corrosion processes, detects the initiation and propagation of cracks resulting from IGA/SCC. The EN signature during SCC results from the creation of new surface area when a crack is initiated or advances. A surge in current occurs as the new surface undergoes the electrochemical processes of dissolution and oxide film formation. The current drops rapidly as the new surface becomes covered with protective oxide. The intensity and frequency of the current and potential pulses are related to crack growth rate. Figure 2a shows typical current and potential noise pulses. Figure 2b shows the standard deviation of the current noise from a tube that developed a through-wall crack in 1,300 hours of superheat using the 40 ppm NaOH feedwater. The standard deviation is in the microampere range during the first 400 hours of the test, suggesting that this is the crack-initiation period. Crack propagation occurs between 900 hours and 1,300 hours of superheating.

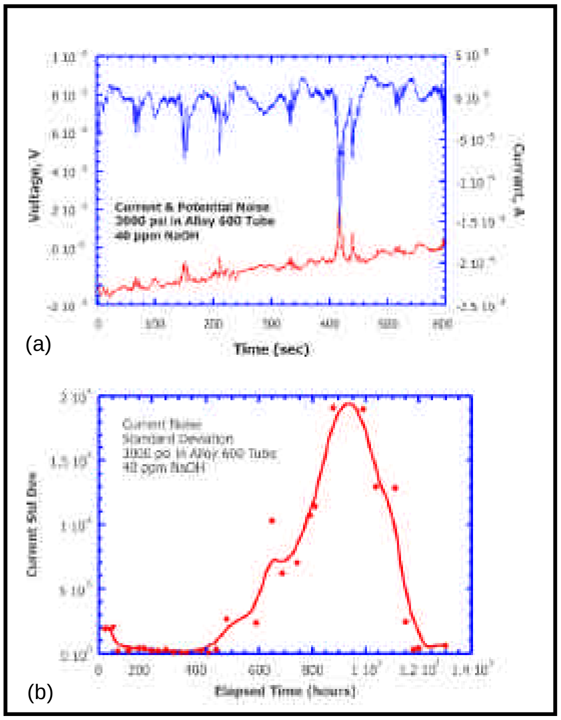



Figure 2. Graph (a) illustrates typical current and potential fluctuations during SCC, and graph (b) plots the standard deviation of the current noise during caustic SCC.

## Planned Activities

The present strategy for mitigating IGA/SCC is based on the assumption that the crack initiation and propagation rate in Alloy 600 steam generator tubes, forming the tube/TSP crevices, depend only on pH and the ECP. Planned work will continue to examine the effectiveness of the present practice of adding hydrazine to the feedwater to inhibit the initiation and propagation of IGA/SCC. The hypothesized rationale for adding hydrazine is that it lowers the ECP of the crevice chemistry to a regime where IGA/SCC does not occur. Preliminary results indicate that hydrazine concentrates in the crevice, and lowers the ECP. However, the ECP values are not sufficiently low to prevent IGA/SCC. The effect of hydrazine concentration in the feedwater and pH have not yet been assessed. The pH effects will be evaluated by controlling the $Na^+/Cl^-$ molar ratio in the feedwater. Computer codes show that the pH in the tube/TSP crevice decreases as the $Na^+/Cl^-$ molar ratio in the feedwater decreases. Crevice chemistry and pH will be determined by chemical analysis of solutions extracted from the crevice. The measured ECP of the freespan and crevice also provide a measure of the pH of the crevice solution. The crevice chemistry results will be used to benchmark the computer codes used by utilities to calculate crevice chemistry and crevice pH. The results of this activity will also provide a preliminary indication of effectiveness of "molar ratio control" as a measure to mitigate IGA/SCC.

Signal analysis and other data analysis procedures will be developed for the EN technique. The analysis will model the transition from microcracks to macrocracks and crack propagation processes. This will enable monitoring of the steam generator crevice for the initiation and severity of IGA/SCC.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Interfacial Transport Phenomena and Stability in Molten Metal-Water Systems

Primary Investigator: M. Corradini, University of Wisconsin-Madison (UW)

Collaborators: Argonne National Laboratory (ANL)

Project Number: 99-233

Project Start Date: August 1999

Project End Date: September 2002

### Research Objectives

A concept being considered for steam generation in innovative nuclear reactor applications involves water coming into direct contact with a circulating molten metal. The vigorous agitation of the two fluids, the direct liquid-liquid contact, and the consequent large interfacial area give rise to very high heat transfer coefficients and rapid steam generation. For an optimal design of such direct-contact heat exchange and vaporization systems, more detailed knowledge is needed relative to the various flow regimes, interfacial transport heat transfer coefficients, and operational stability under reactor-relevant operating conditions. This research project is studying the transport phenomena involved with the injection of water into molten metals (e.g., lead alloys), with the following objectives:

- Design, fabricate, and operate experimental apparatuses that investigate molten metal-water interactions under prototypic thermal-hydraulic conditions,
- Measure the integral behavior of such interactions to determine the flow regime behavior for a range of conditions and stability of these flow regimes,
- Measure the local interfacial mass and heat transfer behavior to ascertain the interfacial area concentration and heat transport length and time scales, and
- Analyze test results to determine an envelope of operating conditions that yields optimal energy transfer between molten metal and water and maximizes stability.

### Research Progress

A comprehensive review of pertinent past experimental investigations has been completed, and new experimental data gained by using different fluid combinations has been compared to some of the past experiments. Two pre-doctoral students and several undergraduate students at the University of Wisconsin - Madison (UW) and two visiting students at Argonne National Laboratory (ANL), working with several scientists and staff at the laboratory, have been instrumental in the fabrication of two large complimentary experimental apparatuses (one at ANL and one at UW). These devices, one of which is illustrated in Figure 1, facilitate the direct contact of water with molten liquid metals, making it possible to carry out detailed studies of the heat transfer and void fraction.

Figure 1. Photograph of liquid metal direct contact heat exchanger facility at the University of Wisconsin - Madison. Tests are conducted with 70 cm of liquid metal in the test section and water injection from 1-10g/s. The facility is capable of running at pressures up to 10bar with liquid metal temperatures at 500°C.

While the two facilities share a common goal, they were designed to provide mutually complementary information. The ANL experiments focus on the heat transfer and flow stability behavior of water injected into molten metal and provide measurements on the evaporation zone length and associated volumetric heat transfer coefficients. The UW experiments focus on two-dimensional mixing behavior and provide real-time X-ray imaging of the multiphase structure of vaporizing water in the molten metal, and have been able to obtain the first of a kind estimates of local average heat transfer coefficients for the compact heat exchanger.

Current experimental results have been discussed during weekly conference calls and several visits among laboratory researchers, resulting in an increased understanding of the physics involved in the direct contact heat transfer between a continuous phase hot liquid at temperatures well above the saturation point of a dispersal injected second liquid. Figure 2 depicts an example of a single frame of high-speed X-ray movies of the void distribution that were obtained with a high-energy X-ray system. The real time X-ray images of this flow allowed the first of its kind measurement of the bubble production time, the bubble rise velocity, and the vaporization rate of water being injected into a high-temperature ( T<400°C) liquid metal pool. These measurements also allowed an estimate of the average local heat transfer coefficient for the vaporization of water in the region just above the nozzle for several different system pressures, injection temperatures, and injection flow rates (Figure 3).

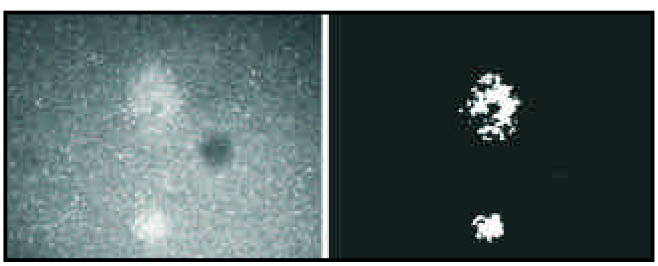


Figure 2. Single frame image of a high-speed movie taken at 100/fps shows water being injected into a liquid metal pool at 500°C. The left image shows a raw image of the bubble formation and evaporation of the water droplets. The right image show the individual bubbles after some image processing. These images can then be analyzed with the aid of a calibration file to determine the local void fraction. From the change in the void fraction the local heat transfer coefficient can be estimated.

The research results from ANL and the UW will constitute a significant database of volumetric heat transfer coefficients and void distribution data associated with direct contact heat transfer, which will aid in the design of advanced heat exchangers for future power

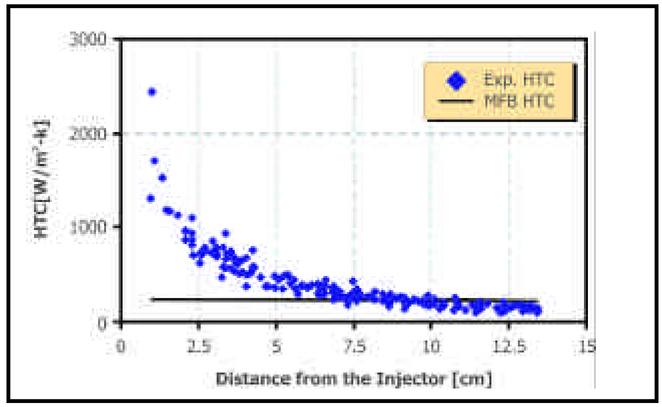



Figure 3. Local average heat transfer coefficients for injection of water into a liquid lead pool at a rate of 1.7g/s at 3.17 bar. The plot shows the average local heat transfer coefficient as a function distance above the water injection nozzle. Above a height of approximately 15 cm, the individual bubbles break up and it is no longer possible to estimate the local heat transfer coefficient. As can be seen, the average heat transfer coefficient approaches the minimum film boiling heat transfer limit (MFB).

reactor designs. The complete set of data from the 1-D and 2-D experiments is currently being assembled for publication in the final research report. The results can be summarized as follows:

(1) Both experiments have obtained similar measurements of the volumetric heat transfer coefficients (in the range of 10 to 20 kW/m$^3$).

(2) This heat transfer rate seems to occur when the injected water is in film boiling within the molten metal pool.

(3) Flow oscillations have been observed at the lower ambient pressures (1 to 2bars) for water injection into the molten metal pool and can be suppressed as the system pressure is increased and to a lesser extent if the water flow is decreased or its temperature approaches saturation.

(4) Dynamic X-ray imaging and associated image reconstruction has allowed measurement of the local void fraction and estimates of the associated local heat transfer coefficients (Figures 2 and 3). These results indicate that the flow regime is bubbly flow with a transition to churn flow at lower pressures. In addition, the local heat transfer coefficient is similar to what may be expected during film boiling heat transfer.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Fundamental Thermal Fluid Physics of High Temperature Flows in Advanced Reactor Systems

Primary Investigator: Donald M. McEligot, Idaho National Engineering and Environmental Laboratory (INEEL)

Collaborators: Iowa State University; University of Maryland; General Atomics; University of Manchester, England; University of Montenegro , Yugoslavia; Kyoto University, Japan; Tokyo University of Science, Japan; Commissariat à l'Energie Atomique (CEA), France

Project Number: 99-254

Project Start Date: August 1999

Project End Date: December 2002

## Research Objectives

This project is a collaborative effort among researchers from laboratories, universities, and industry that couples computational and experimental studies while addressing fundamental science and engineering issues related to new and advanced reactor designs to improve the performance, efficiency, reliability, and enhanced safety of the new reactors, while also reducing their cost and waste levels. This research will provide knowledge in basic thermal fluid science to develop an increased understanding of the behavior of fluid systems at high temperatures, in the application and improvement of modern computation and modeling methods, and in the incorporation of enhanced safety features for nuclear plants. The project promotes, maintains, and extends the nuclear science and engineering base to meet future technical challenges in design and operation of high efficiency and low output reactors, and nuclear plant safety.

INEEL's unique Matched-Index-of-Refraction (MIR) flow system, the world's largest facility of this type, is being applied to obtain, for the first time, fundamental data on flows through complex geometries important in the design and safety analyses of advanced reactors. Successful completion of the study will provide the following new basic science and engineering knowledge:

- Time-resolved data plus flow visualization of turbulent and laminarizing phenomena in accelerated flow around obstructions (spacer ribs) in annuli.
- Application of Direct Numerical Simulation (DNS) and Large Eddy Simulation (LES) for the first time to complex turbulent flows with gas property variation occurring in advanced reactors.
- Fundamental data of internal turbulence distributions for assessment and guidance of Computational Thermal Fluid Dynamic (CFD) codes proposed for advanced gas cooled reactor applications.

## Research Progress

Progress has been on the six tasks:

<u>Heat transfer and fluid flow in advanced reactors:</u> Six areas of thermal hydraulic phenomena have been identified in which the application of CFD techniques can improve the safety of advanced gas cooled reactors. Selection of commercially available CFD codes capable of simulating flows through complicated geometries with large variations of fluid properties has been initiated.

<u>Complex flow measurements:</u> Experimental models were developed for laser Doppler velocimeter (LDV) measurements in the MIR flow system to examine flow in complex core geometries (ribbed annular cooling channels and control rod configurations) and in the transition from cooling channels to formation of jets issuing into a plenum. The initial model was a ribbed annulus forming an annular jet exhausting into the MIR flow system. LDV measurements with this model were completed and were compared to predictions from a commercial CFD code. A second model—a quartz and plastic model that was shown in the 2001 Annual Report—has been fabricated and was assembled in a mockup of the MIR test section. Measurements by LDV and flow visualization have been completed and documented; they are now being analyzed.

DNS development: DNS of laminarizing gas flow and sub-turbulent gas flows have been completed. The turbulent case has been initiated.

LES development: LES results have been obtained for vertical upward flow of air in a channel heated on one side and cooled on another; such a channel flow corresponds closely to the flow in an annular passage with a large radius ratio. This work is the first known LES study of a vertical flow accounting for buoyancy and variations in fluid properties. Work continues on LES codes for heated pipes (Figure 1) and annuli with gas property variation and buoyancy effects. Simulation of flows in annuli with spacer ribs has been initiated. Predictions are in process for comparison to mixed convection measurements in a large rectangular channel at CEA in Grenoble.

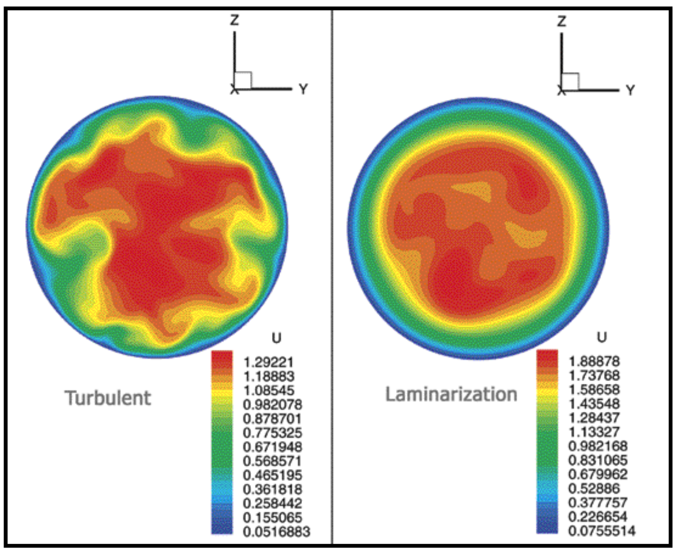



Figure 1. The figure shows large eddy simulations (LES) of strongly-heated gas flow in a vertical tube with buoyancy effects and fluid property variation. Streamwise velocity contours are shown (left = turbulent flow; right = same flow at higher heating rate, causing apparent laminarization, i.e., reduction in turbulent transport).

Miniaturized multi-sensor probe development: Several probes with three and four sensors were developed for application in gases over a velocity range of 0.5 to 15 m/s at temperatures from room temperature to about 1,000K. Three probes were fabricated with three sensors each (one cold-wire, two hot-wires), fitting within a volume of about one mm in diameter. A detailed examination has been conducted of a final probe capable of simultaneous measurement of two velocity components and of temperature in a heated turbulent gas flow, and this probe has been employed in a heated turbulent gas flow in a pipe.

Mixed convection: Data have been obtained to examine the effects of buoyancy forces on heated vertical flows in a pipe, annuli, and a wide rectangular channel. Measurements include mean velocity, temperature, and turbulence profiles as well as wall heat transfer coefficients. These results have been compared to predictions of a research code using a popular turbulence model.

## Planned Activities

In the area of LES development, code development will continue and data comparisons will be made. Calibration procedures and algorithms will be compared and improved, particularly for low velocities. Efforts in mixed convection will include completion of the analysis and documentation of the effects of buoyancy on the turbulent flow of water in a vertical annulus with a heated core. The final technical report will be completed, presenting the key results of all tasks.

NUCLEAR ENERGY RESEARCH INITIATIVE

# An Innovative Reactor Analysis Methodology Based on a Quasidiffusion Nodal Core Model

Primary Investigator: Dmitriy Y. Anistratov, North Carolina State University

Collaborators: Texas A&M University; Oregon State University; Studsvik Scanpower, Inc.

Project Number: 99-269

Project Start Date: August 1999

Project End Date: December 2002

## Research Objectives

The present generation of reactor analysis methods uses few-group nodal diffusion approximations to calculate full-core eigenvalues and power distributions. The cross sections, diffusion coefficients, and discontinuity factors (collectively called "group constants") in the nodal diffusion equations are parameterized as functions of many variables, ranging from the obvious (temperature, boron concentration, and so forth) to the more obscure (spectral index, moderator temperature history, and so forth). These group constants, and their variations as functions of the many variables, are calculated by assembly-level transport codes.

The current methodology has two primary weaknesses that this project will address. The first weakness is the diffusion approximation in the full-core calculation; this can be significantly inaccurate at interfaces between different assemblies. This project will use the nodal diffusion framework to implement nodal quasidiffusion equations, which can capture transport effects to an arbitrary degree of accuracy. The second weakness is in the parameterization of the group constants; current models do not always perform well, especially at interfaces between unlike assemblies. Researchers will develop a theoretical foundation for current models and use that theory to devise improved models. The new models will be extended to tabulate information that the nodal quasidiffusion equations can use to capture transport effects in full-core calculations.

## Research Progress

Progress made on various tasks will be discussed in turn.

Homogenization Methodology for the Low-Order Equations of the Quasidiffusion (QD) Method: A coarse-mesh discretization of the low-order QD (LOQD) equations (Gol'din 1964) was developed that is consistent with the given fine-mesh differencing method for the LOQD equations. It is consistent in the sense that it preserves average values of the fine-mesh scalar flux over the given coarse cells as well as reaction rates, the first and second spatial Legendre moments of the fine-mesh scalar flux over coarse intervals, currents at edges of coarse cells, and the fine-mesh multiplication factor (Anistratov 2002). All these facts are rigorous mathematical results. The definition of discontinuity factors has been derived. The resulting discretization scheme enables one to approximate accurately the large-scale behavior of the transport solution within assemblies. The results from the test problems are extremely encouraging: the coarse-mesh scalar fluxes almost perfectly match every fine-mesh pin-cell average flux, in both the fast and thermal groups.

The developed method can be applied to a general transport method as well, if this method preserves the particle balance. If a fine-mesh solution is obtained directly from a transport differencing method, and it is used to calculate spatially averaged cross sections and special functionals defined in the method, the resulting coarse-mesh solution of the LOQD equations will be consistent with the given transport method. The reason is that the coarse-mesh scheme was derived by algebraically consistent discretization based on the discrete particle balance equation and, thus, this scheme works also for any transport method whose solution satisfies the discrete balance equation.

The developed coarse-mesh algorithm can be coupled with other parts of a complete reactor analysis methodology (e.g., generation of tables of constants, interpolation using tables, pin-power reconstruction).

Improved Boundary Conditions for Assembly-Level Transport Codes: An extension of present-day reactor-analysis methodology was developed that systematically accounts for the effects that different neighbors have on a given assembly's few-group constants (Clarno and Adams 2002). The new technique centers on energy- and angle-dependent albedos that simulate the effect of the unlike neighbors. Each set of albedos defines a branch case and therefore fits into the framework of present-day methodology. The parameter varied in each new branch case is the fractional difference in the neighbor's concentration of an isotope or mixture. (The base case corresponds to a zero difference in all concentrations—an identical neighbor—which produces the usual reflecting boundary condition.) The key simplification is that the albedos are generated by a one-dimensional transport calculation with an homogenized assembly and homogenized neighbor.

The albedo produced from 1D homogenized (1DH) calculations was found to do an extremely good job of capturing the effects of different neighbors in the rather restricted case of lattices that are uniform in one direction (in which the only large-scale variation is in the other direction). In fully 2D problems, the 1DH albedos are accurate near the center of an interface but in general lose accuracy at corners. This loss of accuracy in the albedo produces large errors in corner-pin powers in the worst cases. Very simple modifications to the 1DH albedos have been found to dramatically reduce these large errors. This encouraging result has led the team to pursue systematic (but simple) modifications that are theoretically sound and that produce very accurate results.

The team's complete methodology relies on albedos to estimate the changes in few-group parameters that are induced by differences in a neighboring assembly's composition. Another part of the methodology is to assume superposition and thus build the change in a parameter by summing the partial changes from a variety of differences in a neighbor's composition.

<u>Numerical Method for Solving 2D QD Low-Order Equations:</u> The polynomial-analytic nodal method of Palmtag (1997) was successfully adapted to the solution of the QDLO equations, and the methodology was tested on problems with constant nodal properties (cross sections and Eddington tensor). The QDLO equations are solved for several diffusion test problems (diagonal Eddington tensor with diagonal entries equal to 1/3), and "transport" problems (Eddington tensor with diagonal entries different from one-third and zero or positive off-diagonal components). Several single-node test problems were solved and have reproduced known analytic solutions. A series of two-node MOX-$UO_2$ interface diffusion problems were also solved, and results were compared with reference values and to those calculated by Palmtag (1997). In a representative problem [$UO_2$ (3 percent enriched)-MOX (12 percent enriched)], the fast flux is higher in the MOX assembly, due to its higher fission cross sections. The steepest variation is observed near the surface between the two assemblies, and the flux flattens as the reflecting boundaries are approached. The thermal flux varies strongly at the surface between nodes because thermal absorption is greater in the $UO_2$ assembly than in the MOX assembly. A variety of multi-node (2x2, 3x3 and 4x4) configurations were also solved, and the diffusion results compare favorably with those previously published in the literature.

The transport problems that were solved involve Eddington tensor data in a range observed in homogenized $UO_2$ and MOX assemblies. The increase in values of E increases leakage and therefore decreases k. To more accurately account for transport effects at interfaces between assemblies, the spatial dependence of the homogenized cross sections and Eddington tensor must be incorporated. Equations were derived for a local "correction" to the fluxes generated by the polynomial-analytic nodal method. These equations are designed to yield a zero correction if the diffusion equation adequately represents the physics of the node. If transport effects are present, these finite volume-based correction equations will capture it, to leading-order.

## Planned Activities

The team believes the new methodology is promising, and plans are to continue to refine it, to couple all pieces of a full reactor-analysis system together, and to test the coupled system. This will include the homogenization procedure for 2D assembly-level calculations; method of group constants functionalization using assembly transport solution of 2D multigroup eigenvalue problem with albedo boundary conditions; and numerical method for solving equations of multidimensional coarse-mesh effective few-group nodal QD model, using tables of data parameterized with respect to a set of parameters. The full new methodology must then be compared against the existing state of the art.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Radiation-Induced Chemistry in High Temperature and Pressure Water and Its Role in Corrosion

Primary Investigator: David M. Bartels, Argonne National Laboratory

Collaborators: Atomic Energy of Canada LTD - Chalk River Laboratories

Project Number: 99-276

Project Start Date: August 1999

Project End Date: September 2002

## Research Objectives

Commercial nuclear reactors essentially provide a source of heat that is used to drive a "heat engine" (turbine) to create electricity. A fundamental result of thermodynamics is that the higher the temperature at which any heat engine is operated, the greater its efficiency. Consequently, one obvious way to increase the operating efficiency and profitability for future nuclear power plants is to heat the water of the primary cooling loop to higher temperatures. Current pressurized water reactors (PWRs) run at roughly 300°C and 100 atmospheres of pressure. Designs under consideration would operate at 450°C and 250 atmospheres, i.e., well beyond the critical point of water. This would improve the thermodynamic efficiency by about 30 percent. A major unanswered question is, however, "What changes occur in the radiation-induced chemistry in water as the temperature and pressure are raised beyond the critical point, and what does this imply for the limiting corrosion processes in the materials of the primary cooling loop?"

The direct measurement of the chemistry in reactor cores is extremely difficult, if not impossible. The extreme conditions of high temperature, pressure, and radiation fields are not compatible with normal chemical instrumentation. There are also problems of access to fuel channels in the reactor core. For these reasons, theoretical calculations and chemical models have been used extensively by all reactor vendors and many operators, to model the detailed radiation chemistry of the water in the core and the consequences for materials. The results of these calculations and models can be no more accurate than the fundamental information fed into them, and serious discrepancies exist between current models and reactor experiments. The object of this research program is to generate the necessary radiation chemistry data (yields and reaction rates) needed to accurately model the chemistry in both existing water-cooled reactors, and the higher temperature reactors proposed for the future. This will allow engineers to define the optimal chemical conditions conducive to long life for the primary heat transport system.

## Research Progress

It is impossible in the space of a few pages to even briefly describe all of the results attained on reaction rate measurements in hydrothermal and supercritical water over the past two years. Instead, more details will be provided about what is likely the most important experiments carried out so far. Initial work involved building and testing a cell with which to carry out optical measurements on supercritical water. A comprehensive study of the solvated electron absorption spectrum was carried out, because that is the largest optical absorption available to probe the kinetics of the system. A study was conducted of the reaction of $O_2$ with solvated electrons, as described in the first year report. The kinetics of $SF_6^-$ saturated solutions were measured as described below. The reaction rate of hydrated electrons with perchloric acid was briefly studied, as was the reaction with sodium nitrite and sodium nitrate scavengers. An apparatus was built for high pressure saturation of water with $H_2$ gas. Using this apparatus, measurements of the self-recombination of hydrated electrons were carried out in 100-bar-$H_2$-saturated alkaline water. An extensive series of measurements of nitrobenzene scavenging was carried out; both the reaction with hydrated electrons and the reaction with OH radical were studied. Using the hydroxynitrobenzyl radical absorption as a probe, the reaction rate of OH with $H_2$ has now been measured up to 350°C.

The reaction rate of solvated electrons with $SF_6$ was investigated, because $SF_6$ is to be used to measure the radiolytic yields of solvated electrons. The reaction(s) can be written

$$(e^-)_{aq} + SF_6 \longrightarrow F^- + SF_5 \quad (1a)$$
$$SF_5 + 4H_2O \longrightarrow SO_3^{=} + 5F^- + 7H^+ + OH \quad (1b)$$

The reaction is very specific for hydrated electrons, and tests have shown there is no decomposition of the $SF_6$ under supercritical water conditions. The fluoride ion product can be conveniently measured with ion chromatography. Because acid is a product of the hydrolysis reaction (1b), it was thought prudent to make measurements in alkaline solution to prevent any reaction of electrons with the product acid. In neutral solution, addition of $SF_6$ shortened the electron lifetime, and the signal quickly went to baseline. In alkaline solution, addition of the $SF_6$ accelerates the electron decay at short times, but the signal does not decay to baseline. The signal decays with a second exponential time constant which is independent of the $SF_6$ concentration, but proportional to the hydroxide concentration. The long decay component results from the delayed formation of electrons in the reaction

$$H + OH^- \longrightarrow (e^-)_{aq} + H_2O \quad (2)$$

The rate constants for both reaction (1a) and (2) can be extracted from the biexponential kinetics as a function of temperature and pressure (density). The rate constants determined above the critical temperature at 380°C are plotted against density in Figure 1, along with the rate constants for electron reaction with $O_2$. It is apparent that all three reactions have exactly the same dependence on the density, even though reaction (2) is an order of magnitude slower. The only apparent similarity in all three reactions is that a hydrophobic molecule or atom ($O_2$, $SF_6$, or H) reacts with an anion. It is postulated that this results from a potential of mean force that develops between any hydrophobic species and any charged species in the compressible fluid at intermediate densities. It becomes clear that the radiation induced chemistry in any supercritical-water-cooled-reactor may be very different at the inlet and the outlet of the core.

The detection of reaction (2) in these experiments was a surprise because at lower temperatures the yield of H atom is much smaller than that of electrons. In Figure 2 the ratio $G(H)/G(e^-)$ of the initial radiolytic yields of H atoms and electrons is plotted against the density of the water in various temperature regimes. It is clear that as the density decreases, the H atom becomes favored over the solvated electron. This is ascribed in large part to very fast recombinations of electrons and protons (giving H) at the lower densities, where the enormous reduction in dielectric constant enhances the coulombic attraction. This experimental result shows that H atom chemistry will be much more important in a supercritical water reactor than is the case for PWR water chemistry.

Nitrobenzene was studied to provide a light-absorbing competition partner for other OH radical reactions. The most important of these is the reaction

$$OH + H_2 \longrightarrow H_2O + H \quad (3)$$

of OH with $H_2$, which gives a H atom and water as product. The high pressure $H_2$-saturator apparatus was used to mix $H_2$-saturated water with variable amounts of O2-saturated nitrobenzene solution. Adding the $H_2$

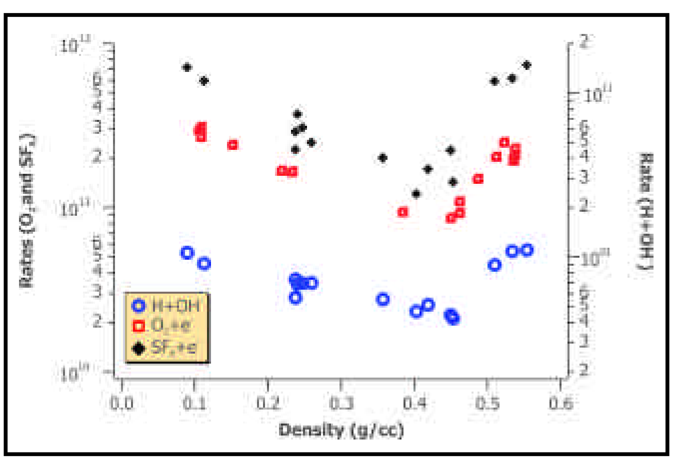



Figure 1. The graphic shows reaction rates for three reactions in supercritical water at 380°C, as a function of the water density. A minimum is seen at about 0.45 gm/cc .

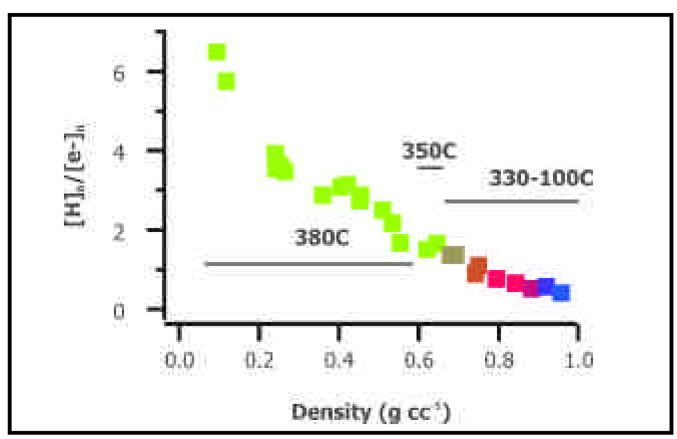



Figure 2. The figure illustrates the ratio of initial concentrations of H atoms and solvated electrons, as a function of the water density in different temperature regimes.

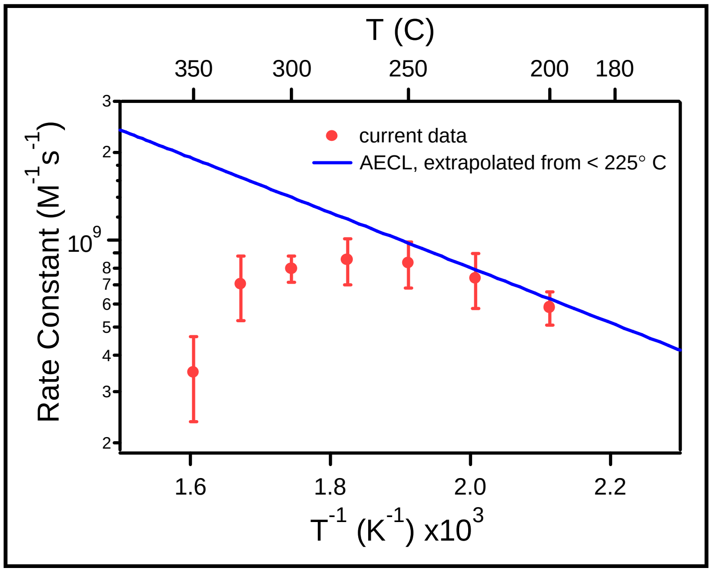



Figure 3. The graph is an Arrhenius plot for the reaction of OH with H2.

intercepts the OH radicals that would otherwise react with nitrobenzene. From straightforward analysis of the absorption growth at 400nm, the rate of the competing reaction can be deduced. The rate constant for reaction (3) from slightly above room temperature to 350°C has now been measured. An Arrhenius plot is shown in Figure 3. The measurements are in agreement with previous work below 250°C. Researchers were astonished to discover that the rate constant for $H_2$ + OH reaches a maximum near 275°C, and then decreases at higher temperature. Unfortunately, the reaction becomes too slow above 350°C for the use of nitrobenzene as the absorbing competition partner. Another scavenger should permit the measurement in supercritical water.

Hydrogen is added to reactor cooling water to inhibit the radiolytic formation of oxidizing species such as $O_2$ and hydrogen peroxide, and to prevent the net radiolytic decomposition of water. Reaction (3) is the key step, because the oxidizing OH radical is converted to the reducing H atom. The amount of hydrogen that is found necessary to inhibit the formation of oxidizers in reactor tests (the critical hydrogen concentration or CHC) is considerably greater than the amount that previous simulations would suggest is necessary. The new reaction-rate result (a lower reaction rate than expected) will explain much of the difference between empirical reactor tests and the predicted CHC. The primary issue is whether the reaction is sufficiently fast to suppress radiolysis in a supercritical water-cooled reactor, with reasonably low $H_2$ overpressure. In all probability this chemistry will still work in supercritical water, but no definitive answer can be given without further experiments.

## Planned Activities

With the end of the funding for this project, it is clear that insufficient data was obtained for a comprehensive model of supercritical water radiolysis. As data for electron recombination is analyzed, a greatly improved model of radiation chemistry in liquid water up to 350°C is expected, which will be of great utility in current PWR calculations. Still missing from the water radiolysis model for reactor chemistry is sufficient information about the chemical yields of neutron radiolysis, which can amount to 30 percent of the total radiation deposited in water. The objectives of a newly funded NERI project are to gather this information, and extend the reaction rate measurements of the current project.

The NERI project has been completed.

# Nuclear Energy Research Initiative

## Novel Concepts for Damage-Resistant Alloys in Next Generation Nuclear Power Systems

**Primary Investigators:** Stephen M. Bruemmer and E.P. Simonen, Pacific Northwest National Laboratory (PNNL)

**Collaborators:** General Electric Global Research & Development (GEGRD); University of Michigan (UM)

**Project Number:** 99-280

**Project Start Date:** August 1999

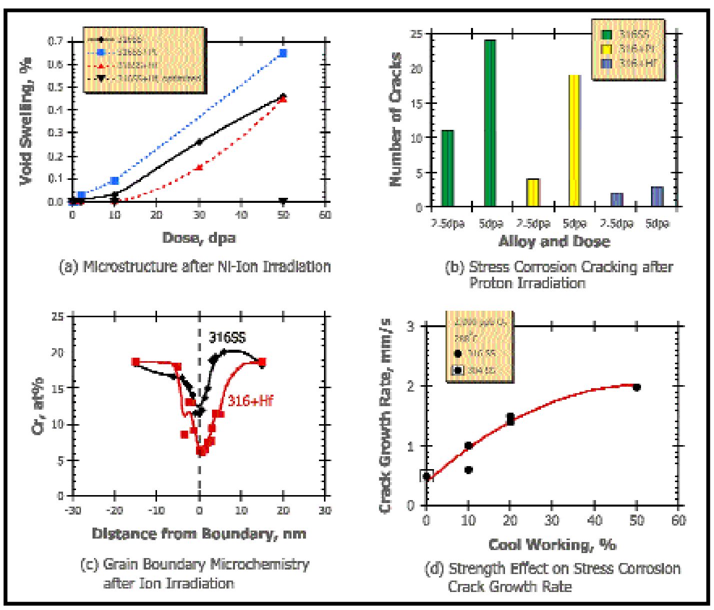
**Project End Date:** September 2002

### Research Objective

The objective of the NERI research is to develop the scientific basis for a new class of radiation-resistant materials to meet the needs for higher performance and extended life in next generation power reactors. New structural materials are being designed to delay or eliminate the detrimental radiation-induced changes that occur in austenitic alloys. These may include a significant increase in strength and loss in ductility (<10 dpa), environment-induced cracking (<10 dpa), swelling (<50 dpa), and embrittlement (<100 dpa). Non-traditional approaches are employed to ameliorate the root causes of materials degradation in current light water reactor systems. Changes in materials design are based on mechanistic understanding of radiation damage processes and environmental degradation, and the extensive experience of the principal investigators with core component response. This work is integrated with fundamental research at PNNL and with focused international projects at PNNL, GEGRD, and UM, led by the Electric Power Research Institute (EPRI). This leveraged approach will help facilitate the revolutionary advances envisioned within NERI. The multi-faceted study of basic and applied science is expected to provide a mechanistic understanding of next generation materials and promote their development. The research strategy capitalizes on the unique national laboratory, industry, and university capabilities that are available for studying radiation damage and an environmental cracking response.

### Research Progress

The highlight of the research is the discovery of an alloy that is resistant to radiation damage based on additions of hafnium (Hf) solute to a low-carbon 316SS. Characteristics and properties of the alloy are illustrated in Figure 1. This damage resistance is supported by characterization of radiation-induced microstructures and microchemistries along with measurements of environmental cracking. Research progress was achieved through a coordinated collaboration among a national laboratory (PNNL - nickel-ion irradiation and characterization); a university (UM - proton irradiation, characterization, and stress corrosion testing); and industry (GEGRD - crack-growth-rate testing in non-irradiated stainless steels tailored to emulate radiation-hardened microstructures). The addition of the oversized element Hf to a low-carbon 316SS reduced the detrimental impact of radiation in contrast to the addition of the




(a) Microstructure after Ni-Ion Irradiation
(b) Stress Corrosion Cracking after Proton Irradiation
(c) Grain Boundary Microchemistry after Ion Irradiation
(d) Strength Effect on Stress Corrosion Crack Growth Rate

Figure 1. The beneficial effects on the characteristics and properties of an alloy when Hf is added to a low-carbon 316SS are shown for (a) void swelling after Ni-ion irradiations, (b) stress corrosion cracking of proton-irradiated alloys, and (c) radiation-induced Cr depletion after high-dose Ni-ion irradiation. These results suggest that the important influence of Hf is on radiation-induced microstructure. The beneficial effect on matrix microstructure is consistent with the (d) strength effect on SCC-growth-rate tests. Hf addition retarded the void swelling shown in (a) and retarded loop formation and hence radiation strengthening.

element platium (Pt), which had no beneficial effects. This reinforces the belief that solute atom size and chemical reactivity are important for enhancing recombination of radiation defects and minimizing damage evolution. Hafnium additions were most effective after specific thermomechanical treatments to maximize the solute content in solution. The presence of Hf in the stainless steel altered microstructure evolution (dislocation loops and voids), altered grain boundary segregation at low doses, and improved resistance to stress corrosion cracking (SCC). Because cracking susceptibility is associated with several material characteristics, separate experiments on non-irradiated stainless steels explored the effects of matrix strength and grain boundary composition on SCC. This work has quantitatively demonstrated the critical importance of matrix strength on crack growth for the first time.

The concept of using oversized solutes to promote catalyzed defect recombination is a major thrust of this NERI project. The successful demonstration of damage resistance in the optimized Hf-doped alloy demonstrates promise for developing damage-resistant alloys for future-generation nuclear reactors. Void formation was delayed in the optimized Hf-doped alloy to radiation doses more than twenty times higher than in the base stainless steel. Resistance to irradiation-assisted SCC measured for proton-irradied samples was also dramatically improved in the Hf-doped alloy. A key next step for assessing this damage-resistant alloy is to evaluate material performance after relevant neutron irradiations. Negotiations were successful with the international Cooperative IASCC Research project to add samples to an irradiation program underway in Russia. Samples from these initial alloys were prepared and are being irradiated to various doses up to ~70 dpa at 330°C.

The collaborative NERI experiments conducted at the three cooperating institutions indicate that radiation effects on strength are more important that radiation effects on grain boundary composition. Radiation-induced strengthening is strongly correlated with susceptibility of stainless steels to IASCC. Strength effects on environmental cracking susceptibility were elucidated on non-irradiated stainless steels after working to systematically increase matrix strength levels. Crack-growth rate increased with strength level for both oxidizing (similar to boiling water reactors) and non-oxidizing (similar to pressurized water reactors) environments. These results suggest that the suppression of radiation-induced grain boundary segregation alone will not assure that an alloy will be resistant to SCC.

The second concept for developing damage-resistant alloys is the use of fine-scale multiphase alloys to mitigate detrimental microstructure evolution during irradiation. Three alloys have been tailored for evaluation of precipitate stability influences on damage evolution. The first alloy was a nickel-base alloy (alloy 718) that was characterized at high irradiation doses for the first time. Microstructural evolution (nanoscale hardening phases and loop/void structures) was examined. The $\gamma''$ phase begins to dissolve at low dose and disappeared altogether at moderate-to-high doses. The $\gamma'$ phase dissolved and re-precipitated, but remained at a small size even to doses of 50 dpa. The experiment demonstrated the benefit of this nanoscale high-density phase to alter loop and void formation and control stability of high-dose irradiated properties. Two precipitation-hardened, Fe-base alloys (PH 17-7 and PH 17-4) were also studied to further evaluate complex second-phase structures on the evolution of radiation damage. One final aspect of evaluating complex multiphase alloys was examined in high-temperature crack-growth tests on specially processed stainless steels. High densities of second-phase carbides at grain boundaries were found to significantly decrease the susceptibility to intergranular SCC. Results demonstrate that proper tailoring of multiphase structures can improve both radiation and environmental damage resistance in stainless alloys.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Advanced Ceramic Composites for High-Temperature Fission Reactors

**Primary Investigator:** R.H. Jones, Pacific Northwest National Laboratory

**Project Number:** 99-281

**Project Start Date:** August 1999

**Project End Date:** September 2002

## Research Objectives

The objective of this research is to develop the understanding needed to produce radiation-resistant SiC/SiC composites for advanced fission reactor applications. The structural and thermal performance of SiC/SiC composites in a neutron radiation field depend primarily on the radiation-induced defects and internal stresses resulting from this displacement damage. The objective of this research is to develop comprehensive models of the thermal conductivity, fiber/matrix interface stress, and mechanical properties of SiC/SiC composites as a function of neutron fluence, temperature, and composite microstructure. These models will be used to identify optimized composite structures that result in the maximum thermal conductivity and mechanical properties in a fission neutron field.

## Research Progress

A newly developed model was used to predict the effects of component (fiber, matrix and interface) parameters and radiation on the overall transverse (through-thickness) thermal conductivity of 2D-SiC/SiC composites. To achieve high overall transverse thermal conductivity, both the matrix and fiber components must have a relatively high thermal conductivity. Furthermore, model predictions indicate that further conductivity enhancement is possible by improving the fiber/matrix (f/m) interface conductance. For instance, while using current interface technology the overall thermal conductivity of a commercially available SiC/SiC composite made with Hi-Nicalon fiber could be about doubled by replacing the Hi-Nicalon with an advanced SiC fiber with higher thermal conductivity (Figure 1, case b). However, by also improving the f/m interface conductance over that available with current design the composite thermal conductivity potentially could be increased by another 25% (case a), i.e., to values exceeding that of stainless steel. For instance, an

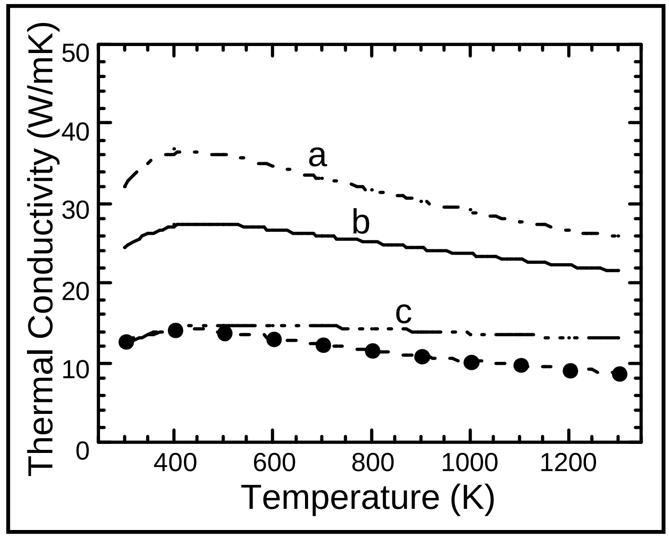



Figure 1. Model predictions of the transverse thermal conductivity for 2D-SiC/SiC composite made with advanced fiber and (a) an advanced f/m interface, (b) current interface technology, and (c) a degraded f/m interface. For comparison, the data points indicate the thermal conductivity for the Hi-Nicalon composite.

oriented graphitic carbon interface might provide such an improvement in interface conductance. However, the thermal conductivity of a composite made with advanced SiC fiber also is more sensitive to degradation effects at the interface. As an example, if only the interface conductance were reduced by a factor of ten (as might happen due to irradiation or other environmental effects) while maintaining the condition of the fiber and matrix components the overall thermal conductivity of the composite with advanced fiber would be reduced by at least 40% (case c). Nevertheless, for such a case the overall thermal conductivity is still about the same as that for the Hi-Nicalon SiC/SiC composite, as indicated in the figure.

A dynamic crack-growth model has been utilized to predict effects of irradiation on the crack growth. Fibers function as bridges that apply closure forces across the crack, which retard crack growth. Thermal- and irradiation-enhanced creep of the fibers reduces the closure force and

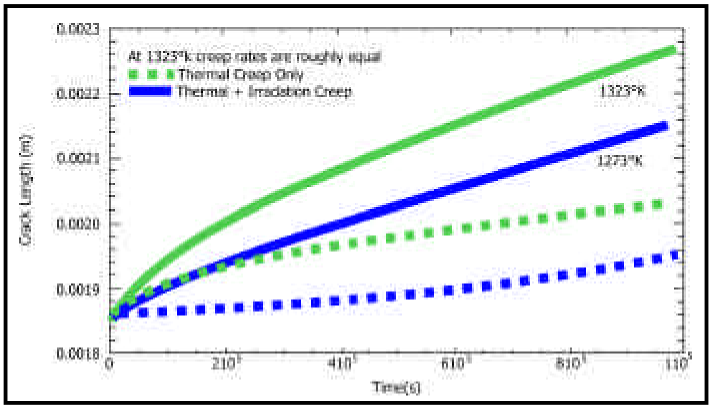



Figure 2. The graph indicates the model result in terms of crack length as a function of time for the indicated test temperatures.

an acceleration in the crack velocity. Experimental data on thermal and irradiation creep of SiC fibers was utilized in the model with the result that thermal creep dominates the growth effects at temperatures above 1,000°C and irradiation creep dominates at temperatures below 1,000°C (Figure 2).

A review of the literature on thermal fatigue and thermal shock of SiC/SiC composites has been completed. Transient thermal conditions will occur in a fission reactor from both the system duty cycle and an accidental loss of coolant. Shut-down of the system will cause some stress build-up in the material with the magnitude being dependent on the cooling rate and thermal gradients. Start-up will also induce stress of the opposite sign to that produced by shut-down and may relax the cool-down stresses. Coolant loss will induce a rapid heating and subsequent rapid stress change. The limited, existing data suggests that continuous fiber ceramic matrix composites such as SiC/SiC exhibit very good thermal shock characteristics, but most data was obtained for $-\Delta T$ conditions as a result of quenching from an elevated temperature. Thermal shock in a fission reactor will result from loss of coolant and will result in a $+\Delta T$. One study was reported for SiC/SiC composites given a $+\Delta T$ with no loss in strength following 25 cycles at a heating rate of 1,700°C/s. Monolithic SiC failed in 1.5 cycles at a heating rate of 1,400°C/s. Thermal fatigue test results also suggest that SiC/SiC composites will exhibit little or no degradation for hundreds of cycles.

The 3-cylinder model was used to calculate the radial, axial, and hoop stresses in the composite components as a function of irradiation dose. An initial residual thermal stress was present due to a $\Delta T$ of -100°C from a nominal synthesis temperature of 1,100°C and an irradiation temperature of 1,000°C. Since previous experimental results indicated that complete fiber-matrix debonding occurred after exposure to low doses at 1000°C for Hi-Nicalon fiber composites, the study monitored the dose dependence of $_{rr}$ at r = r4, the fiber/coating interface. Values of the various domain radii were chosen to match microstructural information for CVI SiC/SiC composites. Total doses were limited to less than 10 displacements per atom (dpa) due to limitations in the swelling data for the various materials.

Large radial stresses are observed to build up at the Hi-Nicalon fiber/carbon coating interface regardless of the properties of the coating, Figure 3. The response of the composite is principally due to the fiber shrinkage during irradiation as the fiber densifies. This is predicted to lead to fiber/matrix debonding at the fiber-coating interface at relatively low doses, which corresponds to the experimental data for this material. Although there is little information on the strength of the interface in radial tension, the model predicts that these stresses approach 1 GPa for a dose of about 1 dpa, which probably exceeds the interface strength. The most important finding is that the radiation-induced shrinkage of Hi-Nicalon leads to fiber/matrix debonding at low neutron doses and, therefore, Hi-Nicalon fibers are unsuited for use as a continuous fiber in SiC/SiC composites exposed to such a radiation field.

Stresses are much smaller for the cases involving the Type-S fiber compared to the Hi-Nicalon fiber. The fiber and the SiC matrix respond identically and there is no differential swelling to contend with for this composite material. Now the response of the composite is principally

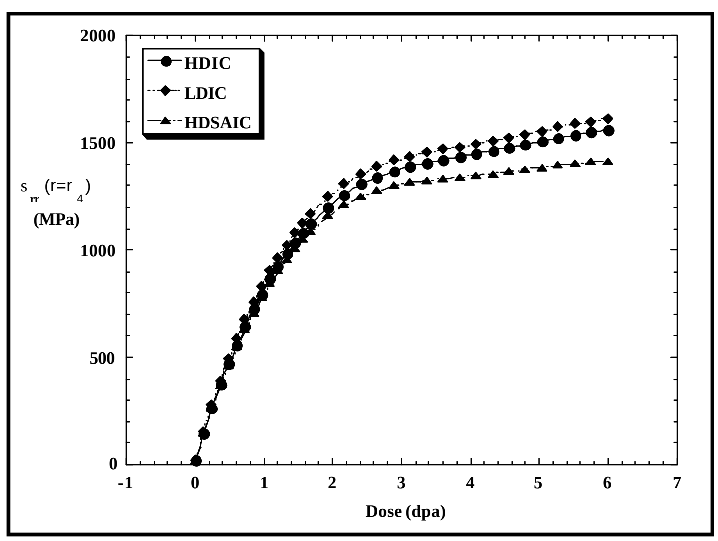



Figure 3. Comparison of three carbon coatings, high-density carbon (HDIC), low-density carbon (LDIC), and high-density slightly anisotropic carbon (HDSAIC) on Hi-Nicalon fibers, showing the radial stresses ( rr) at the fiber/coating interface. The model predicts a minor influence of the coating properties on the radial stress.

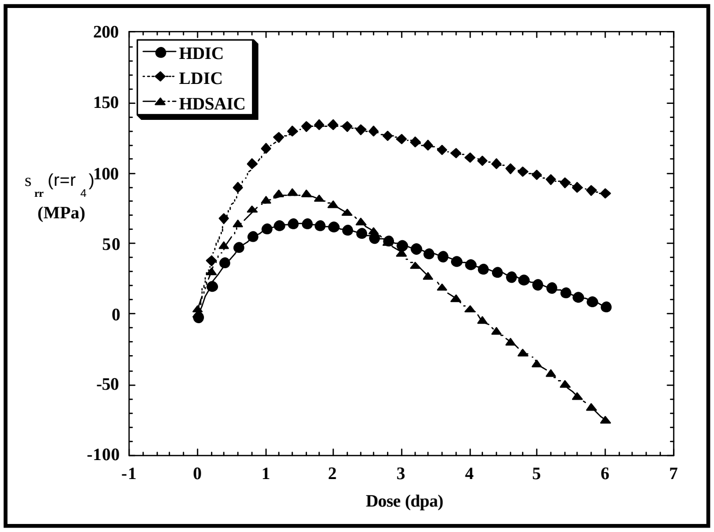



Figure 4. Comparison of three carbon coatings, high-density isotropic carbon, low-isotropic carbon,and high-density slightly anisotropic carbon, on Type-S fibers, showing the radial stresses ( rr) at the fiber/coating interface. The model predicts a significant influence of the coating properties on the radial stress in the system.

due to the radiation response of the carbon coating. Small values of the radial stress at the fiber/coating interface are predicted to lead to retention of fiber/matrix bonding up to significant neutron doses, which corresponds to the experimental data for this material. The most significant finding for the Type-S fiber cases is that the HDIC carbon coating yields the smallest value of the radial stresses in the system, with a peak stress of about 65 MPa predicted after a dose of about 1.5 dpa, Figure 4. The radial stress is then predicted to decrease slowly, with an increasing dose corresponding to the "turn-around" in the HDIC swelling curve. The LDIC material shrinks more than the HDIC and the peak radial stress is predicted to be about 140 MPa. The HDSAIC swells more than either HDIC or LDIC and puts the system into radial compression at the fiber/coating interface. A concern for this coating material is that continued radiation exposure may cause high compressive stresses and interface failure modes that have not yet been observed may be initiated.

Neutron irradiation-induced transmutations result in the presence of both radioactive and stable transmutant elements in the irradiated material. In general, as the neutron irradiation proceeds, the composition of the irradiated material can change due to the addition (or depletion) of solutes and impurities through radiation-induced transmutations, and it can also become radioactive. Activation and transmutation calculations were performed for SiC in the MPBR neutron spectrum. Results for transmutations are shown in Table 1. Calculations were performed for pure SiC irradiated for 10 efpy (4.4 dpa) in MPBR. During that exposure, small concentrations of five elements were produced by transmutation. The concentrations of transmutants are listed in atomic parts per million (appm), and the rate of transmutation relative to damage (appm/dpa) is also shown. For comparison, the transmutation rates per dpa for SiC in HFIR-PTP are also shown, and are very similar to those for MPBR.

Induced radioactivity is generally extremely low for SiC in any neutron environment. In MPBR, after 10 efpy of irradiation and 4 days of cooling, SiC has a residual decay rate of about 4 x $10^{-4}$ Ci/g. This activity is due primarily to 32P,which is a beta emitter. The gamma dose rate at this time is 3 x $10^{-11}$ R/h/cm3 at 1 m due to several short-lived gamma emitters. After cooling a year, the decay rate is 1 x $10^{-6}$ Ci/g dominated by 14C, while the gamma dose rate is 5 x $10^{-14}$ R/h/cm3 at 1 m, due to the long-lived $^{26}$Al produced from Si by a two-step reaction.

As reported earlier, dpa cross sections for SiC as a function of neutron energy have been developed under this project as reported earlier. With these cross sections, the dpa for SiC irradiated in a particular facility can be determined by integrating the DPA cross sections with the neutron energy spectrum of the facility. This procedure has been used to

| | MPBR | | HFIR-PTP |
|---|---|---|---|
| Element | Concentration (appm) | appm/dpa | appm/dpa |
| P | 36 | 8.2 | 6.1 |
| H | 8.0 | 1.8 | 3.3 |
| He | 5.8 | 1.3 | 2.5 |
| Mg | 3.6 | 0.8 | 1.5 |
| Be | 1.5 | 0.3 | 1.4 |

Table 1. Concentrations of elements in SiC produced by neutron irradiation induced transmutations after irradiation in the MPBR for 10 full power years (4.4 dpa). The transmutation per dpa is compared with that for SiC in HFIR-PTP.

| | | DPA/efpy | |
|---|---|---|---|
| Reactor | Position | Fe | SiC |
| MPBR | core/He coolant | 0.46 | 0.44 |
| BWR | midplane | 2.8 | 2.4 |
| PWR | midplane | 3.7 | 3.1 |
| HFR | C5 | 12 | 11 |
| ATR | midplane | 14 | 12 |
| EBR-2 | midplane | 25 | 27 |
| HFIR | PTP mid | 33 | 28 |
| FFTF-MOTA | midplane | 43 | 53 |

Table 2. Displacement damage rates in dpa per effective full power year (DPA/efpy) for SiC in several fuel and materials test reactors, commercial reactors, and a pebble bed reactor design. Values for pure Fe are shown for comparison to typical metals (listed in order of increasing values for SiC).

generate spectrally averaged displacement cross sections for SiC in a number of reactors that are or have been used for radiation damage testing of structural nuclear materials, in typical commercial reactors, and in the Modular Pebble Bed Reactor (MPBR). Calculations of DPA damage rates were also made, and the results are listed in Table 2 for several reactor positions. The damage rate for MPBR is less than one-fifth of that for the commercial pressurized water reactor (PWR) and boiling water reactor (BWR). Test reactors have damage rates more than an order of magnitude greater than the 0.44 dpa/efpy for MPBR, so they could quickly irradiate test materials to MPBR lifetime doses.

## Planned Activities

The NERI project has been completed.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Isomer Research: Energy Release Validation, Production, and Applications

Primary Investigator: John A. Becker, Lawrence Livermore National Laboratory (LLNL)

Collaborators: Los Alamos National Laboratory (LANL); Argonne National Laboratory (ANL)

Project Number: 00-123

Project Start Date: April 2000

Project End Date: September 2003

## Research Objectives

The goal of this applied nuclear isomer research program is the search for, discovery of, and practical application of a new type of high energy density material (HEDM). Nuclear isomers could yield an energy source with a specific energy as much as a hundred thousand times as great as that of chemical fuels. There would be enormous payoffs to the Department of Energy and to the country as a whole if such energy sources could be identified and adapted to a range of civilian and defense applications. Despite the potential payoff, efforts in applied isomer research have been rather limited and sporadic. Basic research on nuclear isomers dates back to their discovery in 1935 with an occasional hint of further progress since then to tantalize interest in HEDM. In most cases, these hints were refuted following careful examination by other groups.

The isomer research area is rich with possibilities and several areas were prioritized as likely to be the most rewarding and fruitful for initial experimental and theoretical investigation. These areas bear directly on important issues: Can the energy stored in nuclear isomers be released on demand? Is the size of the atomic-nuclear mixing matrix element large enough to be useful? Under what circumstances? Can quantal collective release of isomeric energy be initiated from a Mössbauer crystal? What is the precise energy of the 3.5 eV level in $^{229m}$Th?

Specific experiments have been targeted to provided some answers:

- X-ray induced decay of isomeric Hf ($^{178m2}$Hf) with a sensitivity $10^5$ times that of recent work
- NEET: A measurement of the atomic-nuclear mixing matrix element in $^{189}$Os
- Stimulated emission in isomeric Te ($^{125m}$Te)
- Superradiance in isomeric Nb ($^{93m}$Nb)
- Energy and lifetime of the $^{229m}$Th isomeric level at 3.5 eV
- TEEN: Nuclear isomer energy release in isomeric Hf $^{178m2}$Hf

## Research Progress

Triggered decay of a nuclear isomer is clearly one requirement for usefulness of isomers as an energy source. Research in the past two-years focused on the question, "What is the cross section for keV X-ray induced decay of the 31-y isomer in the nucleus $^{178}$Hf with nuclear excitation energy 2.4 MeV?" The question is relevant because induced decay had been reported in isomeric Hf $^{178}$Hf with an integrated cross section of $10^{-21}$ cm$^2$-keV, orders of magnitude greater than nuclear cross sections (Collins et al., 1999, Phys. Rev. Lett. 82, 695, and Collins et al., 2002, Europhys. Lett. 57, 677). This team (Ahmad et al., 2001, Phys. Rev. Lett. 87, 072503) has reported an upper limit approximately 5 orders of magnitude below that of Collins (1999) for $E_X$>20 keV (see Figure 1). The 2002 report of Collins et al., claims that the induced decay occurs at lower X-ray energies than they previously reported (near 10 keV). This work was done at the Japanese 3rd Generation Synchrotron light source, SPring-8.

The collaborative team from ANL, LANL, and LLNL believes this result is also specious. They designed and fielded a second experiment at the ANL Advanced Photon Source (APS) in 2002 with an experimental arrangement optimized for low energy X-ray bombardment, but still taking advantage of the intense "white" beam as opposed to utilizing an monochromatic beam. This arrangement permits more photons incident on the target than in an

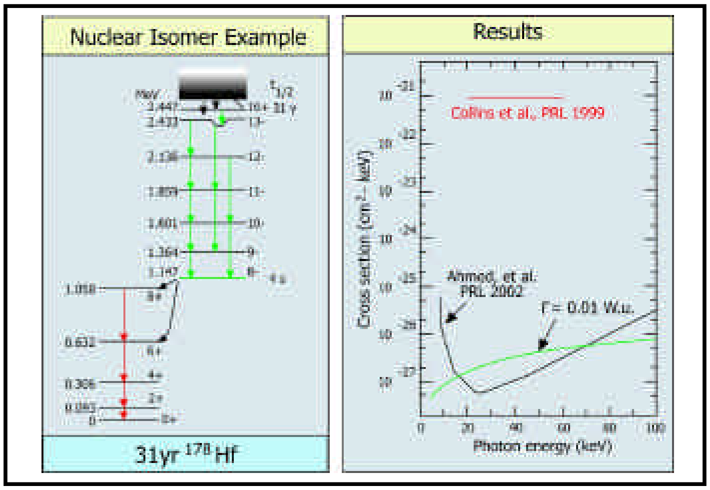



Figure 1. The graphs illustrate the decay scheme of 31-y isomeric Hf-178. The cross-section results of Ahmad et al. (2002) and Collins et al. (2001) are shown on the right-hand side. The green line shows the cross section expected for E1 absorption to a level at $E_{photon}$ with strength 0.01 W.u. as a function of $E_{photon}$. The strength of the photon absorption (0.01 times a single particle unit) represents a reasonable upper limit to the transition strength based on nuclear

experiment with a quasi- monoenergetic beam, without "holes" in the incident X-ray flux. Thin Hf targets enriched in the 31-y isomer mounted on Be disks were used in the experiment. On-line analysis suggests that the cross section $\sigma_{X\text{-}ray}$ for induced X-ray emission is less than $10^{-27}$ $cm^2$-keV at the 5 σ limit for $E_X$>6.5 keV. This result is orders of magnitude below claims made any positive published claims.

## Planned Activities

Work will move on from the isomeric Hf issue to attack the physics of nuclear isomers relevant to their use as an energy source. The particular focus will be on the following areas:

- Documentation of the experiments on the X-ray induced enhanced decay of isomeric $^{178}$Hf, with a focus on low incident X-ray energies. Controversial experimental reports continue to circulate in the literature and in the community of scientists working in isomer physics (see above).
- NEET, Nuclear Excitation by Electronic Transition, in $^{189}$Os. The process has been demonstrated in $^{197}$Au, in a recent synchrotron experiment in Japan. The matrix element is similar to the (inverse) internal conversion matrix element. Important conditions for NEET to compete with real photon emission will include an energy overlap of the atomic and nuclear states, and a common multipolarity of the atomic and nuclear transitions. An experiment is being developed to prepare ionized atomic $^{189}$Os by bombardment with a variable energy electron beam in an electron beam ion trap (EBIT) and to pump a nuclear state in $^{189}$Os at 216.6 keV. The energy of the electron beam is carefully controlled and tuned so that the sum of the energies of the bombarding electron beam and the L-shell ionized $^{189}$Os (a free-bound transition) add up to the excitation of the nuclear $^{189}$Os level at 216.6 keV. Trapped ions are periodically gathered up and counted. The signal is the energy and decay rate of the $J^{\Pi}=9/2^-$, $E_X$=30.814 keV, $t_{1/2}$=5.7 h state, populated in the decay of the 216.6-keV nuclear state. The experiment is sited at LLNL's EBIT facility.
- Stimulated emission of isomeric $^{125m}$Te. Large quantities of isomeric $^{125m}$Te enable an experiment to demonstrate stimulated emission of photons from nuclear isomers. The signal is the enhancement of time-correlated photons observed in a solid state detector located on the axis of a rod-shaped Mössbauer source. The Khlopin Radium Institute in St. Petersburg, Russia, has developed a program (under ISTC auspices) to extract $^{125}$Sb from spent nuclear fuel as a generator of 20 percent $^{125m}$Te. The content of $^{125}$Sb in 20 to 50 GW-day fuel after a four-to six-year cooldown is 3 curies per kilogram of uranium. This is a larger source of isomeric $^{125m}$Te than previously available. A 0.5-cm long source of magnesium tellurate containing 20 percent $^{125m}$Te would produce a stimulated gamma ray at the rate of 3.9 x $10^{-3}$/sec, observable over accidental coincidence rate 8 x $10^{-4}$/sec. If successful, this would be the first observation of stimulated emission of gamma rays. In order to achieve gain, Borrman channeling or some other effect requiring single crystals would be necessary to reduce the attenuation of 109-keV gamma rays. The Khlopin Radium Institute has been contacted to determine the availability and schedule the acquisition of $^{125m}$Te. Once the source is acquired, a magnesium tellurate Mössbauer source will be made, and the experiment fielded. The needed apparatus to observe time-correlated photon emission exists in the laboratory at Los Alamos and can be set up within a few months.
- Superradiance in isomeric $^{93m}$Nb. Superradiance is an effect discovered by Robert Dicke in the 1950's that results in an enormous broadening of the photon channel through the cooperation of an ensemble of quantum excited states. The possibility of nuclear

superradiance was recognized for Mössbauer crystals in the 1960s. One of the most likely candidates for exhibiting nuclear superradiance is $^{93m}$Nb. The observation of enhanced photon emission would be the first evidence of the broadening of the photon channel width necessary to exploit the stored nuclear energy in a nuclear isomer. Molybdenum isotope production targets in the Medical Isotope Program are a source of $^{93m}$Nb at LANSCE. Approximately, 300 grams of molybdenum was processed last year to extract 0.5 milligrams of niobium. This brings the total inventory at LANL to 3 milligrams. The inventory of $^{93m}$Nb is now 0.2 milligrams. An attempt will be made to create a single crystal containing a high enrichment of $^{93m}$Nb from the present stock. The synthesis of potassium heptaflouroniobate crystals is being attempted from Nb-HF-KF aqueous solution. This method is being examined as a possible method for producing single crystals containing 93mNb. A search will be made for enhanced photon emission along nuclear Bragg angles following successful crystal growth.

- $^{229}$Th ground state doublet (3.5 eV). What is the energy of the first excited state of $^{229}$Th near 3.5 eV? The uncommonly low energy of the ground state doublet would allow a laboratory isomer that could be manipulated by a laser. There are also potential applications as a radionuclide thermal source (RTG). Researchers at LLNL and LANL are working with colleagues at ANL to establish the feasibility of such an experiment, taking advantage of unique facilities at ANL.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Random Grain Boundary Network Connectivity as a Predictive Tool for Intergranular Stress-Corrosion Cracking

Primary Investigator: Mukul Kumar, Lawrence Livermore National Laboratory

Collaborators: University of Michigan; General Electric Corporate Research & Development

Project Number: 01-084

Project Start Date: October 2001

Project End Date: September 2004

## Research Objectives

Intergranular stress corrosion cracking (IGSCC) is one of the most pervasive degradation modes in current light water reactor systems and is likely to be a limiting factor in advanced systems as well. In structural materials, IGSCC arising from the combined action of a tensile stress, a "susceptible" material, and an "aggressive" environment has been recognized for many years and the mechanisms widely investigated. Recent work has demonstrated that by sequential thermomechanical processing, properties such as corrosion, IGSCC, and creep of materials can be dramatically improved. The improvements have been correlated with the fraction of so-called "special" grain boundaries in the microstructure. A multi-institutional team comprising researchers from Lawrence Livermore National Laboratory (LLNL), University of Michigan (UM), and General Electric Corporate Research & Development (GECRD) propose an alternative explanation for these observations: that the effect of grain boundary engineering is to break the connectivity of the random grain boundary network through the introduction of low-energy, degradation-resistant twins and twin variants. A collaborative science and technology research project is being carried out that is aimed at verifying the mechanism by which sequential thermomechanical processing ameliorates IGSCC of alloys relevant to nuclear reactor applications, and prescribing processing parameters that can be used in the manufacture of IGSCC-resistant structures.

In this work, methods are being developed to quantify the interconnectivity of the random grain boundary network and measure the interconnectivity of a series of materials where it has been systematically altered. Property measurements are then performed on the materials, their performance ranking compared with the boundary network measurements, and the materials characterized to correlate actual crack paths with the measurements of the random grain boundary network.

With this data, the methods that have been chosen to describe the random grain boundary network will be evaluated and improved. The characterization method will be tested by evaluating the interconnectivity of the random grain boundary network in a series of as-received materials, their expected performance ranked, and that result compared with property measurements.

The major accomplishments of this project are expected to be (1) the determination that the random boundary network connectivity (RBNC) is a major driver of IGSCC in low to medium stacking fault energy austenitic alloys, (2) the development of a predictive tool for ranking IGSCC performance of these alloys, and (3) the establishment of thermomechanical processing parameters to be applied in the manufacture of IGSCC resistant materials. The outcome of the project will be identification of a mitigation strategy for IGSCC in current Light Water Reactor (LWR) conditions that can then enable the development of economically and operationally competitive water-cooled advanced reactor systems.

## Research Progress

The experimental test material, Inconel 600 of nominal composition Ni-16Cr-9Fe (by weight percent) was subjected to cycles of sequential thermomechanical processing. Each processing cycle consisted of rolling at room temperature to a reduction in thickness of 25 percent, annealing at 1,025°C for 18 minutes (following a 42-minute heating ramp), and water quenching. This cycle was repeated up to four times, so specimens were analyzed in a total of five processing conditions (including the as-received state).

Specimens for electron back-scatter diffraction (EBSD) were sectioned to analyze the microstructure at the center of the rolled and annealed sheets, in order to avoid artifacts associated with the specimen surfaces. Metallographically prepared specimens were examined in a Hitachi S2700 or Philips XL30S scanning electron microscope, both with an automated EBSD attachment from TSL, Inc. (Draper, Utah). Orientation information was acquired on a hexagonal grid, with total scan areas in the range 1.1 x $10^5$ to 2.5 x $10^6$ $\mu m^2$, using a step size in the range 0.5-5 $\mu$m.

The analysis of EBSD data was carried out using custom algorithms written using Interactive Data Language software from Research Systems, Inc. (Boulder, Colorado). Grain boundaries were categorized according to the Brandon criterion, using 2° as a minimum disorientation for a low-angle boundary. Those boundaries with $\Sigma \geq 29$ were considered special, including low angle and coherent twin boundaries. The number fraction ($f_n$) of special grain boundaries was determined for each specimen from populations of more than 1,000 individual boundaries.

The changes in the grain boundary network structure was quantified using new cluster analysis algorithms developed for this project. The two-dimensional network of grain boundaries determined from EBSD data was analyzed by identifying boundary clusters, each cluster consisting of all the interconnected boundaries of like type. In this work, the focus is on clusters of only random or only special boundaries. Clusters were identified using a depth-first graph-search algorithm. About 632 grains were analyzed for each specimen. Figure 1 shows the mass fraction, $m_s$, for clusters of size s in Inconel 600, as a function of processing history. The cluster mass is the total (dimensionless) length of boundary contained in the cluster. In percolative systems, cluster masses tend to be distributed across several orders of magnitude, particularly near the percolation threshold. Therefore, the data in Figure 1 has been collected in bins spaced evenly in log(s); the size s on the x-axes in this figure represents the upper bound of each bin. Physically, Figure 1 indicates what fraction of the total length of boundaries in the specimen is occupied in clusters of size s. For example, Figure 1a pertains to the random boundary clusters in the as-received material, where the majority of boundaries are incorporated into a single large cluster of mass 391 units (i.e., spanning hundreds of grains).

In Figures 1a-e, the effect of grain boundary engineering on random boundary clusters is shown quantitatively. After just one cycle of processing (Figure 1b), the largest interconnected clusters of random boundaries are broken up, and the largest clusters have mass in the range 100-178 units. On each subsequent processing cycle (Figures 1c-e), the random boundary network becomes increasingly fragmented, and larger populations of small clusters emerge. After four processing cycles, all of the clusters have mass below 32 units, more than an order of magnitude smaller than the largest cluster mass in the as-received condition. Although there has been considerable discussion surrounding the connectivity of random grain boundaries during grain boundary engineering, investigators believe that these results are the first direct quantitative measurement of such connectivity and its evolution as a function of processing history. The dramatic fragmentation of the random boundary network documented in Figure 1 may explain the concomitant remarkable improvements observed in material properties after grain boundary engineering.

Figures 1f-i show the mass distributions for clusters of special grain boundaries, and therefore represent the complement to Figures 1a-e for the random boundary clusters. In the as-received state, special clusters are extremely small and isolated (Figure 1f). With each cycle of processing, the connectivity of the special boundaries improves, and after four processing cycles, the largest fraction of boundaries is of mass 100-178 units. Compared with the as-received material, this is an order of magnitude increase in the maximum cluster mass. Thus, the order-of-magnitude decrease in random cluster mass is symmetrically offset by a 10-fold increase in the mass of special clusters.

The foregoing discussion can be simplified by considering simple scalar measures of the cluster mass, such as the maximum cluster mass and the weighted average mass. These two parameters are shown in Figure 2 as a function of the processing cycle number for both random and special clusters. Both the average and maximum cluster mass exhibit similar trends, reflective of the construction of contiguous special boundary networks at the expense of large random boundary clusters, which rapidly break into much smaller clusters during processing. Although the special clusters grow at the same rate that the random clusters fragment, they do not grow to the very large masses that the random clusters exhibit in the as-received state.

The cluster mass changes illustrated in Figure 1 are accompanied by changes in the length scales of the clusters. The decrease in random cluster size and the corresponding increase in special cluster size during sequential thermomechanical processing are shown in Figure 3. The maximum linear cluster dimension, $D_{max}$, represents a projection of the largest contiguous path of random or special boundaries in the two-dimensional section, while the correlation length ξ is a representation of the average diameter of all clusters measured in a given specimen. As expected from the trends observed in cluster mass (Figures 1, 2), the process of grain boundary engineering leads to a significant reduction in the random boundary cluster size, by about a factor of three.

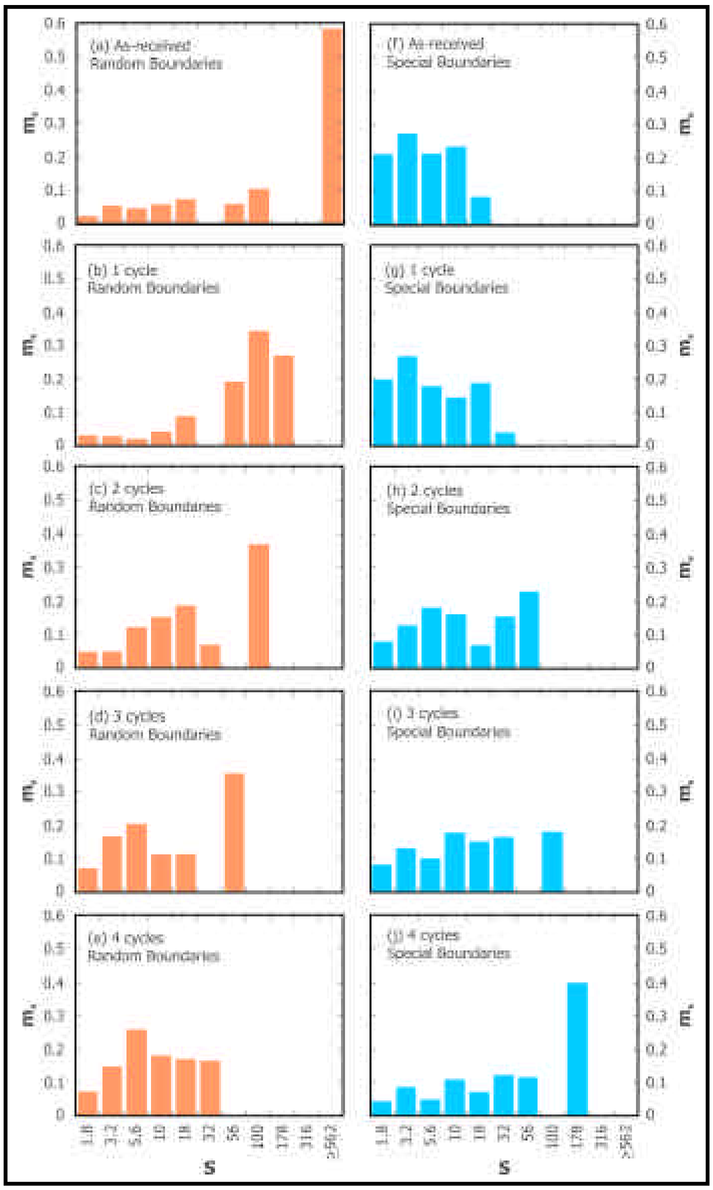



Figure 1. The graphs are a quantitative depiction of the changing grain boundary network topology during grain boundary engineering: (a)-(e) show the cluster mass distributions for only the random boundaries after 0,1,2,3, and 4 cycles of processing, respectively, and (f)-(j) show the complementary mass distributions for the special boundary clusters.

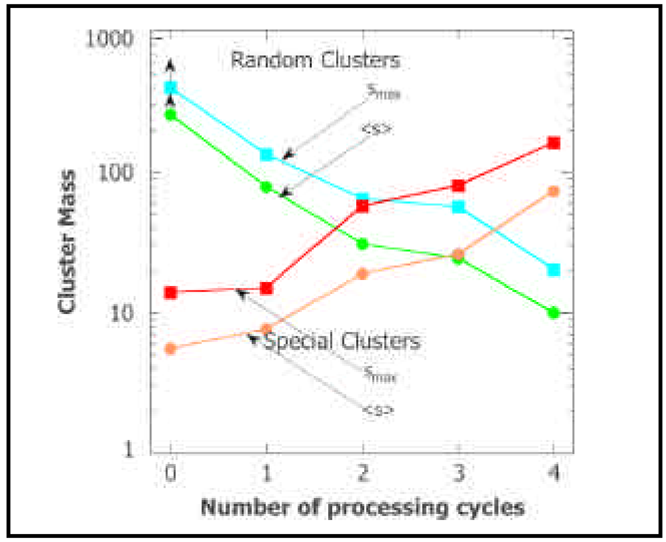



Figure 2. Changes in grain boundary cluster masses are plotted as a function of grain boundary engineering processes. Both the maximum cluster size ($s_{max}$) and the weighted average mass (<s>), are shown for random and special clusters. Random clusters for the as-received material extended beyond the scan area, so the data points represent a lower-bound.

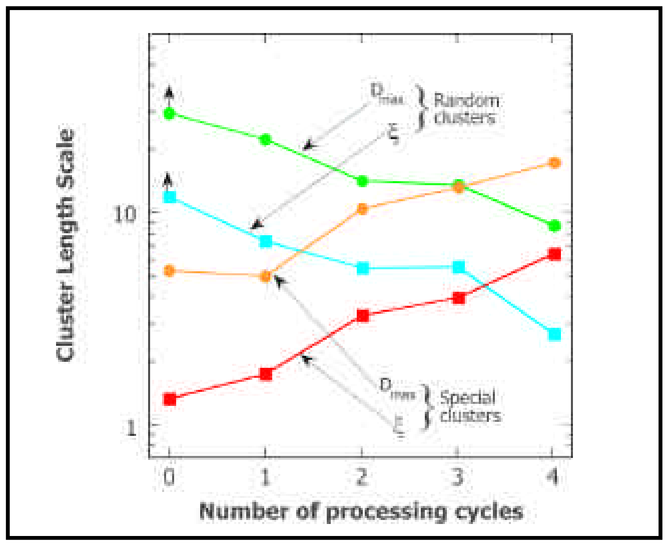



Figure 3. Changes are illustrated in the characteristic length scales of grain boundary clusters during grain boundary engineering, including the maximum cluster dimension ($D_{max}$) and the correlation length ($x$) for random and special boundary clusters.

The experimental test material, 304 grade stainless steel, was subjected to cycles of sequential thermomechanical processing. Each processing cycle consisted of rolling at room temperature to a reduction of 20 percent and annealing at 1,000°C followed by water quenching. This cycle was repeated up to four times with an annealing time of 60 minutes after the first rolling pass and then for the subsequent cycles these times were sequentially reduced by 10 minutes. The specimens were analyzed in the as-received state, and after two and four processing cycles. A further heat treatment was done after the fourth cycle for an hour at 1,000°C to get a grain size comparable to the as-received condition, but this had no effect on the other microstructural parameters.

Specimens for electron back-scatter diffraction (EBSD) were sectioned to analyze the microstructure at the center of the rolled and annealed sheets, in order to avoid artifacts associated with the specimen surfaces. Metallographically prepared specimens were examined in a Philips XL30S scanning electron microscope with an automated electron backscatter diffraction (EBSD) attachment from TSL, Inc. (Draper, Utah).

The analysis of EBSD data was carried out using custom algorithms written using Interactive Data Language software from Research Systems, Inc. (Boulder, Colorado). Grain boundaries were categorized according to the Brandon criterion, using 2 as a minimum disorientation for a low-angle boundary. Those boundaries with 29 were considered special, including low angle and coherent twin boundaries. The number fraction ($f_n$) of special grain boundaries was determined for each specimen from populations of more than 1,000 individual boundaries.

The results are given below in Figures 4a and 4b. The random grain boundaries in all cases are ones in black and the colored boundaries in the background are crystallographically special boundaries. The as-received condition is to be used as the baseline microstructure, as its properties are well understood. The sample after four cycles of thermomechanical processing and additional heat treatment has a lower fraction of special boundaries as well as a more connected network of random grain boundaries. It is anticipated that this will provide a good comparison in the IGSCC behavior.

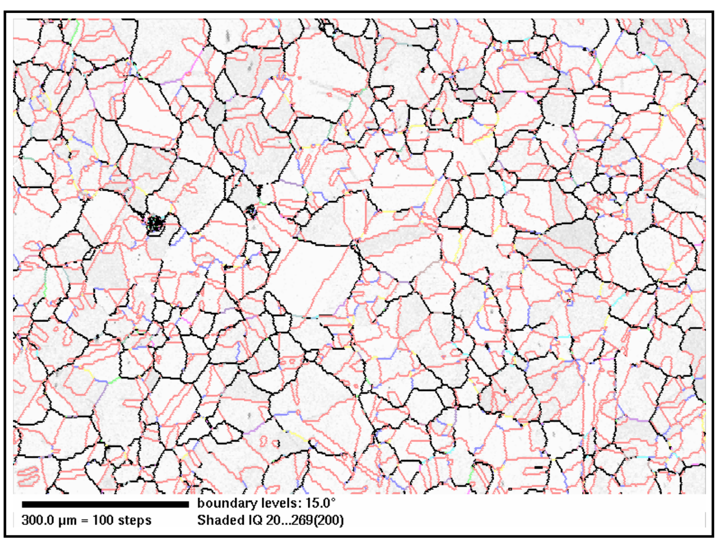



Figure 4a. The as-received condition microstructure is to be used as the baseline.  (Special fraction: By length (fL) = 0.65; By frequency (fN) = 0.53.)

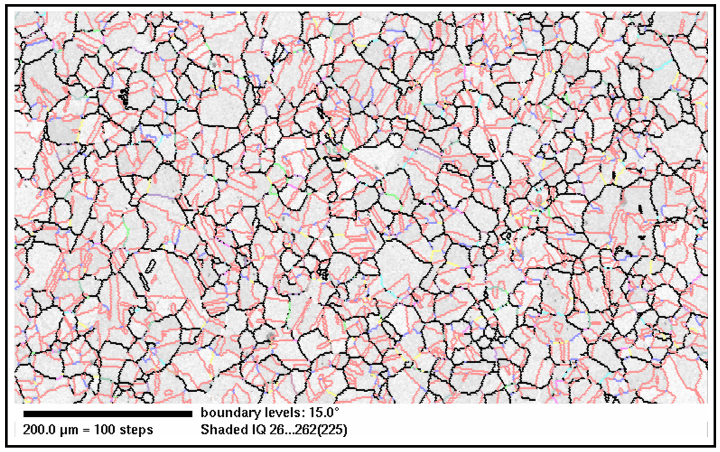



Figure 4b. The microstructure is pictured after 4 cycles of thermomechanical processing.  (Special fraction: By length (fL) = 0.53; By frequency (fN) = 0.35.)

## Planned Activities

The susceptibility of the microstructure to crack initiation will be assessed using constant extension rate tensile (CERT) testing. CERT tests will be conducted in pressurized water reactor (PWR) primary water (320°C - 360°C, containing a hydrogen overpressure) or in boiling water reactor (BWR) water (288°C - 350°C, with varying corrosion potential and water purity) and a strain rate of $3 \times 10^{-7}$ $s^{-1}$. Samples will be periodically removed from the autoclave and examined for intergranular (IG) cracking. The CSL-type of boundaries on which IG cracks appear will be determined by reference to the grain boundary character distribution recorded prior to the IGSCC test. In this way, a strain-dependent map of grain boundary cracking and a tally of the types of grain boundaries that crack will be developed. Crack depth can also be measured using an acetate replica technique.

Following completion of the test, samples will be cross-sectioned and polished, and the grain boundaries along the crack path will be characterized to determine the propensity for cracking of each grain boundary type. The grain boundaries comprising each triple point will also be analyzed.

The Stress Corrossio Cracking (SCC) growth rate response for different microstructures will be compared under different water chemistry conditions. These experiments are performed using fracture mechanics specimens (0.5T or 1T CT specimens) in sophisticated equipment involving precision control and monitoring of water chemistry and corrosion potential; digital servo-control loading systems under computer control; high pressure high temperature autoclave systems with digital temperature controllers; continuous high resolution direct current potential drop monitoring of crack length; and computer data acquisition and test control.

Tests will be performed at about 25 ksi√in in 288°C high purity water containing 2,000 ppb $O_2$ or 100 ppb $H_2$ to represent typical boiling water reactor and pressurized water reactor conditions, respectively. On completion of testing, the specimen will be fatigued apart and evaluated by scanning electron microscopy.

Further plans for FY 2003 include processing of 304 stainless steel and Inconel 600 to optimize the microstructure by increasing the special fraction. These efforts are underway.

Testing to compare the IGSCC behavior will be conducted in the first quarter of FY 2003.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Reactor Physics and Criticality Benchmark Evaluations for Advanced Nuclear Fuel

Primary Investigator: William J. Anderson, Framatome ANP, Inc. (FANP)

Collaborators: Sandia National Laboratories (SNL); Oak Ridge National Laboratory (ORNL); University of Florida (UF)

Project Number: 01-124

Project Start Date: September 2001

Project End Date: September 2004

## Research Objectives

The objective of this Nuclear Energy Research Initiative (NERI) project is to design, perform, and analyze critical benchmark experiments for validating reactor physics methods and models for fuel enrichments greater than 5-weight (wt) percent $^{235}U$. These experiments will also provide additional information for application to the criticality-safety bases for commercial fuel facilities handling greater than 5-wt percent $^{235}U$ fuel. Because these experiments are to be designed not only as criticality benchmarks but also as reactor physics benchmarks, investigators wish to include measurements of critical boron concentration, burnable absorber worth, relative pin powers, and relative average powers.

## Research Progress

The first task of this project was to identify the fuel to be used. Options considered were to use fuel from existing stockpiles, or to fabricate new fuel. In addressing this issue, the project identified a significant stockpile of 6.93-wt percent enriched $UO_2$ located at Pennsylvania State University. This fuel, the "Pathfinder fuel," consists of 417, unirradiated, 6-fuel rod assemblies. Although the fuel is much longer than the critical facility can use, the fuel is suitable for refabrication into shorter rods. The use of this fuel for this project provides the desired enrichment for these experiments, and eliminates a major stockpile of enriched uranium from Pennsylvania State University's regulated inventory.

Having identified the fuel, the next major task was to identify a suitable shipping container for transporting the fuel from Pennsylvania State University to Sandia National Laboratories. This shipping package needed to be licensed to contain up to 7.0-wt percent-enriched fuel. The container also had to be long enough to accommodate the 82.5-inch long Pathfinder elements. Because no suitable container was identified with a license to contain up to 7.0-wt percent-enriched fuel, licensing a shipping package became a major challenge.

The first option investigated was the MO-1 shipping package. This was selected because the Department of Energy's Oak Ridge Operations Office had agreed to be shipper of record and already owns two MO-1 shipping packages. However, the MO-1 had to be eliminated as a viable option because it is a "grand-fathered" package that has not been drop tested to current standards.

Attention was then focused on the modified WE-1 shipping package, a Type A shipping package that was recently modified by adding an armor plated steel inner container. The addition of this inner container has made it a Type B shipping package suitable for shipping various higher-enriched uranium fuels. A safety analysis of the WE-1 containing the Pathfinder fuel is underway, which involves the following steps:

- Designing an inner container to hold the Pathfinder fuel in the WE-1
- Performing a criticality safety evaluation to demonstrate that the WE-1 Pathfinder fuel shipment will meet regulatory requirements for criticality safety
- Performing a structural/stress analysis to demonstrate that the WE-1 Pathfinder fuel shipment will meet regulatory requirements
- Submitting a license amendment to the NRC for review

On March 31, 2002, the safety analysis was on schedule for to be submitted to the NRC on May 15, 2002.

Another task in Phase 1 was to design the experiments to be performed at Sandia National Laboratories. The preliminary design configuration involves water-moderated, square-pitched lattices of cylindrical fuel rods containing enriched $UO_2$ pellets (approximately 6.93-wt percent $^{235}U$). The arrays consist of fully flooded, uniform lattices of aluminum-clad fuel rods.

The Preliminary Design Report, issued in February 2002, describes four suites of fully flooded critical configurations with square-pitch lattices. The experiments consider two pitch values, 0.800 cm and 0.855 cm. Due to the uncertainty of fuel re-fabrication costs, two base designs are documented. These consist of a 3x3 array of 15x15 assemblies, and a cruciform array of 17x17 assemblies. Although the flexibility of the 3x3 array of 15x15 assemblies makes it the preferred design, the cost of constructing 1,836 fuel rods may require that the cruciform array of 17x17 assemblies, which uses 1,596 fuel rods, be used.

In support of future experiments, SNL is preparing a safety basis for the experimental facility, the SPRF/CX. The existing authorization basis (AB) for the SPRF/CX was written to cover the activities associated with the BUC-CX (the BUC-CX is a test of the reactivity effects of fission-product poisons on a critical system). The current experiment differs from the BUC-CX in the following ways:

(1) The mass of fuel in the current experiment is higher than is authorized for the BUC-CX.

(2) In the final configuration, the reactivity worth of the soluble poison will be considerably higher than the allowed excess reactivity.

(3) The experiments involve irradiation to produce sufficient fission products in the fuel to allow the relative fission density to be measured as a function of fuel rod location. The fission products are part of the source term for the accidents considered in the AB. The increased inventory needs to be accounted for in the AB update.

(4) If experiments at elevated temperatures are to be done, consideration will have to be given to the reactivity effects of the elevated temperatures and the impacts of any cooling that might occur and result in positive reactivity insertion.

The foregoing list suggests several points for consideration in the SPRF/CX AB update, given the test matrix for the current experiment.

The design of the experiments will incorporate the results of a sensitivity/uncertainty (S/U) analysis. The S/U analysis is being performed to determine how well commercial reactor fuel with $^{235}U$ enrichments greater than 5-wt percent fit into the area of applicability of existing critical benchmark experiments. Second, the S/U tools are being applied to the proposed experimental design to ensure that it is optimized to provide the best possible coverage for the commercial fuel of interest.

A review of existing critical benchmarks identified a total of 154 experimental configurations with $^{235}U$ enrichments in the range of 5 to 10 wt percent. Sensitivity data for 125 of these experiments have been generated with the SEN3/KENO analysis sequence. Sensitivity data have also been generated for eight configurations of the proposed experimental design. Critical boron searches were performed for each configuration, and then the sensitivity data were generated for the eight critical configurations.

Part of this project considers the impacts of higher enrichments on fuel fabrication and handling for commercial facilities. Reviews of two of the three major commercial fuel processing and fabrication facilities in the United States indicated that some fuel processing operations are convertible to higher enrichment operation with acceptable cost increases. However, some large batch operations appear to be more challenging with regard to higher enrichments. The primary problem area from a cost standpoint may be the packaging and transportation of $UF_6$ cylinders.

To date, all the tests to be performed by Framatome ANP, Inc. and ORNL are on schedule, or ahead of schedule. SNL efforts have been delayed due to work on the BUC-CX program. Over the next two quarters, SNL is expected to accelerate its schedule and complete its tasks within the current schedule.

The research subcontract with the University of Florida was not signed until February 2002. Although this placed UF behind schedule on all tasks, work has commenced and UF is expected to complete all tasks on schedule.

## Planned Activities

The remainder of Phase 1 includes the following tasks:

- Completion of the sensitivity/uncertainty analysis (ORNL)
- Documentation of the Final Design Report (FANP)
- Analysis of the experiments using industry codes (FANP, ORNL, UF)

- Revisions to the SNL AB (SNL)
- Completion of criticality safety, stress, and containment analysis for the WE-1 (FANP)
- Submittal of the WE-1 license amendment to the NRC (FANP)
- Detailed investigation of $UF_6$ packaging and transportation considering >5 wt percent enriched $UF_6$ (UF)
- MCNP scoping calculations to review the sizing of critically safe processing equipment considering >5 wt percent enriched fuel (UF)

NUCLEAR ENERGY RESEARCH INITIATIVE

# Fundamental Understanding of Crack Growth in Structural Components of Generation IV Supercritical Light Water Reactors (SC LWR)

Primary Investigator: Iouri Balachov, SRI International

Collaborators: VTT Manufacturing Technology

Project Number: 01-130

Project Start Date: August 2001

Project End Date: September 2004

## Research Objectives

The objectives of this project are as follows:

- Increase understanding of the fundamentals of crack growth in structural components of Generation-IV supercritical light water reactors (LWRs) made of stainless steels and nickel base alloys at supercritical temperatures.
- Provide tools for assessing the influence of the operating conditions in power plants with supercritical coolant temperatures on the electrochemistry of different types of corrosion processes taking place in the coolant circuits of supercritical power plants.
- Measure material-specific parameters describing the material's susceptibility to stress corrosion cracking and other forms of environmentally assisted degradation of structural materials at supercritical coolant conditions.
- Use these measurements to interpret the rate-limiting processes in the corrosion phenomena and as input data for lifetime analysis.
- Use the SRI-developed FRASTA (fracture surface topography analysis) technique to obtain information on crack nucleation times and crack growth rates via analysis of conjugate fracture surfaces. Identify candidate remedial actions by which the susceptibility to stress corrosion cracking can be decreased.

A unique combination of two advanced techniques is used for studying material reliability. Controlled Distance Electrochemistry (CDE) allows investigators to determine in relatively short experiments a measurable material parameter that describes the transport of ions or ionic defects in the oxide films. This will be correlated with the susceptibility to cracking, using FRASTA to reconstruct the evolution of crack initiation and growth.

## Research Progress

Progress made in examining the fracture surfaces will be discussed.

Crack behavior in the specimens tested in the environment is not easy to interpret, and it is frequently difficult to establish a correlation with loading conditions. A commonly used method of determining the crack length, the DC potential drop measurement, is very useful in providing continuous reading the crack tip position during the test. However, such a method requires careful calibration when applied to a new geometry; it also gives an averaged value through the thickness and through the fracture process zone.

Significant information related to crack behavior can be obtained by examining the fracture surfaces. This section will report the results of applying the SRI-developed fracture surface topography analysis (FRASTA) technique. The FRASTA technique characterizes the topography of conjugate fracture surfaces, juxtaposes conjugate fracture surface profiles on the computer, and assesses the inelastic deformation that occurs at the crack tip during crack extension by manipulating the spacing between the conjugate surfaces. Using the inelastic deformation on the fracture surface, it is possible to reconstruct the crack growth history and microfracture processes and present the results graphically.

The fracture surface topography was characterized by SRI's FRASTAscope, which consists of a confocal-optics scanning laser microscope, a computer-controlled precision x-y stage, and computer software to control the system and create the topography data files.

The FRASTA technique was applied to one of the samples, a single-edge-notch and wedge-loaded disc specimen. This specimen was fatigue pre-cracked and

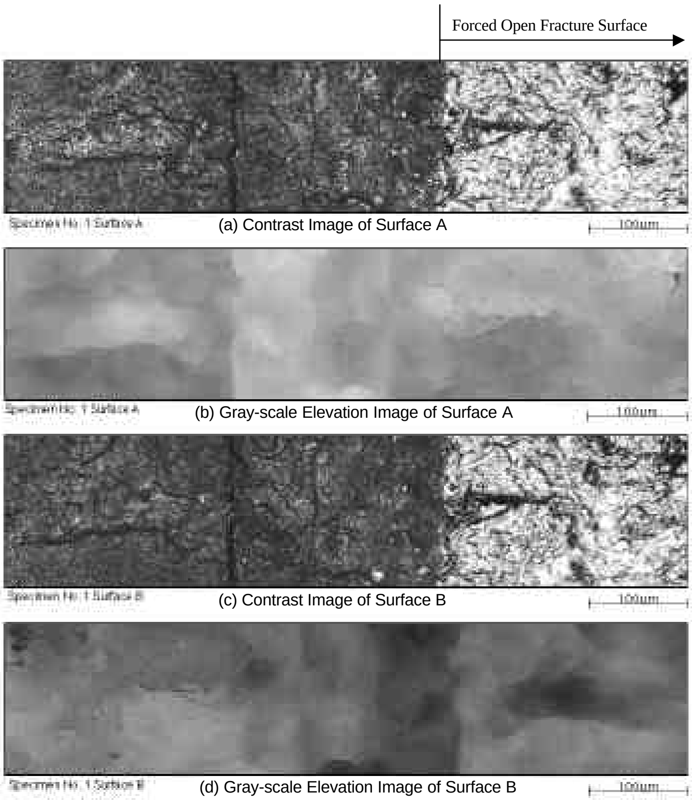



Figure 1. The graphics show contrast and gray scale topography images of conjugate surfaces.

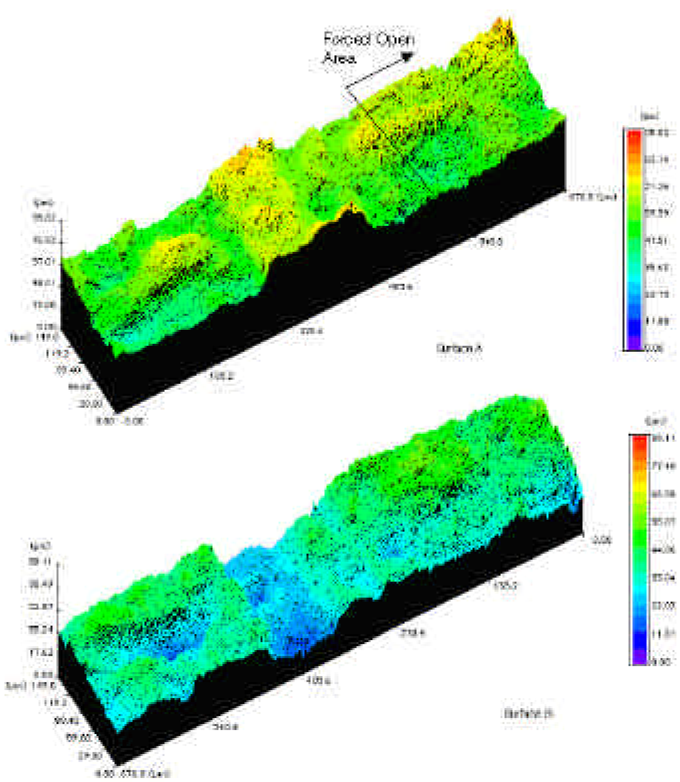



Figure 2. The images provide a perspective view of fracture surfaces.

wedge-loaded at room temperature, then placed in an autoclave to expose it to a supercritical water environment. After several interrupted exposures, the specimen was removed from the autoclave and the crack was opened by applying cyclic loading.

Figure 1 shows the contrast and gray-scale topography images of the area covering the end of the fatigue pre-crack, crack growth during the test in the environment, and the beginning of forced opening after the test. In the gray-scale topography images, white areas represent high elevation and dark areas low elevation. Images of Surface B were flipped horizontally for easier comparison with Surface A. The areas exposed to the environment were covered with oxide film and appear dark. Forced-open areas were highly reflective surfaces. In the dark area in the vicinity of a forced-open fracture surface, several vertical markings were observed. It is difficult to determine where the crack growth in the environment started through the visual examination of the SEM fracture surface images. Thus, researchers are interested in using FRASTA analysis to determine the significance of these markings and also the precise location of the transition from the fatigue pre-crack to the growth of the crack in the supercritical environment.

Figure 2 shows a perspective view of the topography of the fracture surfaces. Several steps in elevation mentioned above are more clearly seen. These steps are parallel to the crack front.

The fracture process was reconstructed using the topography information. Some of the results are shown in the fractured area projection plots (FAPPs) of Figure 3. A well-defined crack front is not seen in the FAPPs. One of the reasons for this is that there are some errors in the elevation data; thus, when conjugate surfaces were matched, the crack front became a zone rather than a single line. The magnitude of elevation error in this analysis is not negligible because the surface reflection was relatively poor due to use of oxide film, and the degree of deformation was small.

Although the crack front was not well-defined, there were some trends in the crack front movement through the series of FAPPs. However, the more precise trend in the crack front movement could be characterized by constructing the fractured area (white area) as a function of conjugate surface spacing. The results are shown in Figure 4.

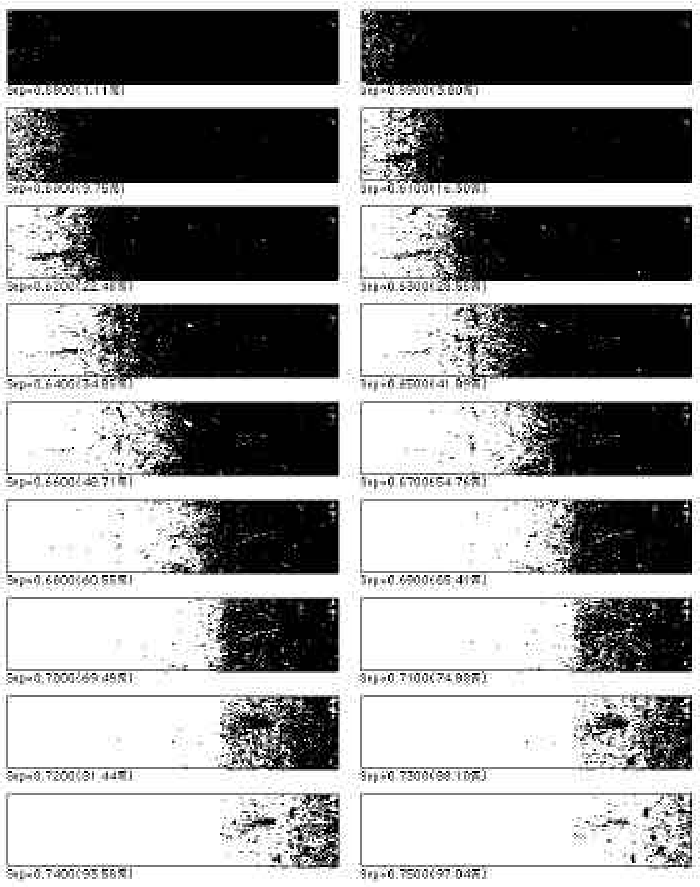


Figure 3. Pictured above are a series of FAPPS showing the crack growth processes. The plots show hesitation in crack front movement at some locations.

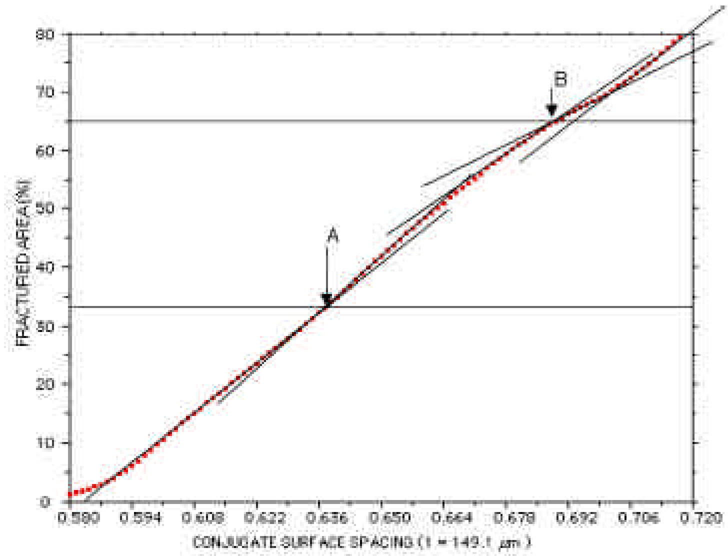



Figure 4. The graph illustrates the fractured area increasing the slope of the curve as a function of conjugate surface spacing.

The curve shows that the slope was approximately constant up to Point A, indicating that the condition of crack growth up to this point was steady and unchanged. However, at Point A, the slope changed and became a little steeper. A steeper slope suggests that the material was less resistant to the crack growth. Thus, some change occurred at Point A that changed the characteristics of crack growth. A possible reason for this could be the change of loading conditions-the change from fatigue pre-cracking at room temperature to a supercritical water environment. The state at Point A will be examined in detail later. The change observed at Point B is due to the forced opening of the crack. It was shown later that the crack front position observed in the FAPP at Point B corresponds to the location where surface color changed, as seen in Figure 1. The slope becomes significantly less steep after Point B, suggesting that the crack growth in this region required more plastic deformation than that in the environment.

## Planned Activities

Two parallel activities are planned for years two and three of the project: (1) Electrochemical studies of properties of the oxide films and (2) fracture surface analysis for candidate structural materials.

Tasks scheduled for "An Electrochemical Characterization of the Oxide Film Properties on Candidate Materials in Supercritical Conditions in the Bulk and within the Crack" follow:

- Characterize the formation and reduction kinetics and the properties of metal oxide films by using the Contact Electric Resistance (CER) technique.
- Measure the solid contact impedance spectra of oxide films by using Contact Electric Impedance (CEI).
- Characterize the oxidation and reduction kinetics and mechanisms of metals as well as the properties of metal oxide films by using Thin-Layer Electrochemical (TLEC) impedance measurements.
- Perform CDE measurements in simulated crack conditions. The influence of adding ionic species/contaminants on the oxide film formed on the surface of candidate materials will be explored experimentally in a static autoclave under simulated conditions corresponding to those existing inside a stress corrosion crack.

- Rank the influence of ionic species on the oxide films forming on metal surfaces exposed both to simulated supercritical LWR bulk coolant and to simulated crack chemistry conditions, using advanced electrochemical techniques.
- Quantify and interpret the rate-limiting processes in the corrosion phenomena and the role of electrochemical reactions and properties of oxide films in the crack growth mechanism under supercritical conditions.
- Derive material-specific parameters that describe the susceptibility of metals to stress corrosion cracking in supercritical coolant conditions for the materials in which crack growth is controlled by the phenomena in oxide films within the crack.
- Identify candidate remedial actions (changes in water chemistry, material chemical composition, and metallurgical parameters, in particular the degree of cold work) that can decrease the susceptibility to stress corrosion cracking.

Tasks in the "Experimental Investigation of Crack Initiation and Propagation and Estimation of Life-Time of Candidate Structural Materials" are as follows:

- Expose loaded specimens to a supercritical aqueous environment simultaneously with electrochemical studies of the oxide films on the same materials.
- Examine fracture surfaces of specimens (they will be broken after the tests) by using the FRASTA technique.
- Identify crack nucleation sites and times for specimens made of different candidate materials.
- Use FRASTA to determine crack front formation and movement, including formation of discontinuities ahead of the crack and their possible coalescence later while the crack is advancing.
- Estimate crack growth rates for different candidate materials under a set of conditions using FRASTA for examination of conjugate fracture surfaces.
- Correlate electrochemical information on material-environment interactions with crack nucleation and growth data from FRASTA to delineate the fundamentals of uniform and localized degradation of structural materials in Generation-IV supercritical LWRs and estimate life-times of candidate materials for structural components under a variety of normal

NUCLEAR ENERGY RESEARCH INITIATIVE

## New Design Equations for Swelling and Irradiation Creep in Generation IV Reactors

Primary Investigator: Wilhelm G. Wolfer, Lawrence Livermore National Laboratory

Collaborators: Pacific Northwest National Laboratory; Lawrence Berkeley National Laboratory

Project Number: 01-137

Project Start Date: October 2001

Project End Date: September 2004

### Research Objectives

The objectives of this research project are to significantly extend the theoretical foundation and the modeling of radiation-induced microstructural changes in structural materials used in Generation IV nuclear reactors, and to derive from these microstructure models the constitutive laws for void swelling, irradiation creep, and stress-induced swelling, as well as changes in mechanical properties.

The need for the proposed research is based on three major developments and advances over the past two decades. First, new experimental discoveries have been made on void swelling and irradiation creep that invalidate previous theoretical models and empirical constitutive laws for swelling and irradiation creep. Second, recent advances in computational methods and power make it now possible to model the complex processes of microstructure evolution over long-term neutron exposures. Third, it is now required that radiation-induced changes in structural materials over extended lifetimes be predicted and incorporated in the design of Generation IV reactors.

The approach in this effort to modeling and data analysis is a dual one in accord with the objectives to both simulate the evolution of the microstucture and develop design equations for macroscopic properties. Validation of the models through data analysis is therefore carried out at both the microscopic and the macroscopic levels. For the microstructure models, results were used from transmission electron microscopy of steels irradiated in reactors and from model materials irradiated by neutrons as well as ion bombardments. The macroscopic constitutive laws will be tested and validated by analyzing density data, irradiation creep data, diameter changes of fuel elements, and post-irradiation tensile data. Validation of both microstructure models and macroscopic constitutive laws is a more stringent test of the internal consistency of the underlying science for radiation effects in structural materials for nuclear reactors.

### Research Progress

A microstructure code was developed that evaluates the full, time-dependent distribution of vacancy clusters and the dislocation density, all within a mean-field framework. The vacancy cluster distribution ranges from monomers to voids of arbitrary size. The stochastic, atomic processes of void growth or shrinkage are included by master equation and Fokker-Planck treatments of the void size distribution function. Void growth and fluctuation rates are determined from a self-consistent calculation of the thermal and radiation-induced vacancy and interstitial monomer populations. The dislocation subsystem is modeled entirely as network dislocations, in terms of a single density parameter. Interstitial aggregation is not allowed, as dislocation loops are not explicitly considered. However, it will be included in the future. Dislocation bias factors are calculated according to previously described methods (Sniegowski and Wolfer 1983). There is a cutoff distance used in the derivation that is related to the average density of dislocations.

In the research, two choices are made for that parameter. The first is to calculate dislocation bias factors assuming a density of $6 \times 10^{14}$ $m^{-2}$, a typical terminal density under steady irradiation. This is the procedure in Wehner and Wolfer (1985), where it would be expected to give the experimentally observed asymptotic swelling rates for calculations with a fixed dislocation density. The second choice is to calculate the bias factors using a cutoff obtained from the instantaneous value of the evolving dislocation density. That density is evolved according to a model (in Wolfer and Glasgow 1985) that incorporates dislocation-dislocation annihilation processes along with a dislocation multiplication from pinned dislocations undergoing irradiation-driven, biased climb. The model includes one free parameter-namely, the mesh length or pinning density (set to a value of 400 nm to fit the observed terminal dislocation densities in irradiated steels), which is determined by the density of carbide precipitates in the steel.

It was verified that this team's implementation of the dislocation evolution model reproduces earlier results for annealing in 316 stainless steel. Likewise, the combined void plus dislocation simulations were checked to verify they conserve mass, both with and without irradiation. The method is computationally efficient to temperatures of 650°C, at which point the stable void size becomes too large for efficient simulations. For all lower temperatures, the simulations extend to much longer times than previous treatments of stochastic void nucleation. This method is applied to a model of a high-purity, type-316 austenitic stainless steel.

It is easy to see a temperature-dependent, incubation-like period in the simulated void swelling versus time. Volumetric swelling curves are presented in Figure 1 for a series of temperatures (340°C to 540°C) for an irradiation dose rate of $10^{-6}$ dpa/s and an initial dislocation density of $6x10^{13}$ m-2. The data shown in Figure 1 is obtained using a constant dislocation bias factor, as in Wehner and Wolfer (1985). The values for 316 stainless steel are 1.63 for interstitials and 1.04 for vacancies. The simulations show a brief incubation period, which increases in duration at lower temperatures. Subsequently, the swelling rates at different temperatures are comparable, around 0.85 percent/dpa. This asymptotic rate is largely dictated by the dislocation bias factors. Overall, the predicted swelling behavior is similar to experimental data, both in the appearance of an incubation-like feature and in terms of the numerical value of the asymptotic slope. However, the duration of the incubation period is short compared to commercial steels, being more similar to irradiation results obtained for pure binary and ternary austenitic steels.

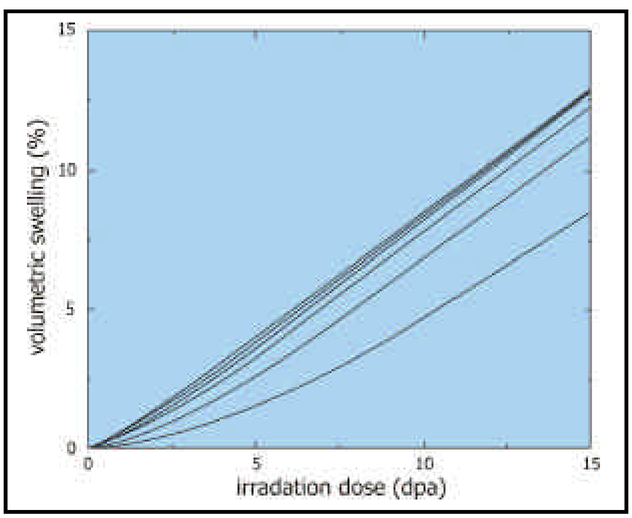



Figure 1. The graph plots volumetric swelling, $D$V/V in percent, versus total irradiation dose for a pure, type-316 stainless steel. The dose rate is $10^{-6}$ dpa/s; the starting dislocation density is $6x10^{13}$ $m^{-2}$. The various curves correspond to temperatures of 340°C to 540°C in increments of 40°C. The 340°C curve has the longest incubation period. While the dislocation density is allowed to evolve with time, the dislocation bias factors derived from the stress-induced interaction with the vacancy and interstitial defects are taken to be constant.

The swelling predictions are very sensitive to the choice of dislocation bias factors. Figure 2 displays the swelling curves using dislocation bias factors that are calculated according to the instantaneous dislocation density. The predicted dislocation bias factors are found to be smaller at low dislocation densities, for example 1.39 and (1.03) for interstitials (vacancies) at a dislocation density of $6x10^{13}$ $m^{-2}$, and they increase with dislocation density. This gives rise to a prolonged period of void nucleation in solution-annealed metals, producing a striking, incubation-like behavior that lasts from 2 to 15 dpa of total fluence. Besides lengthening the incubation period, the density-dependent bias also causes the asymptotic rates of swelling to depend noticeably on temperature. This change in behavior occurs because the terminal dislocation density is temperature-dependent, the calculated dislocation bias factors are now density-

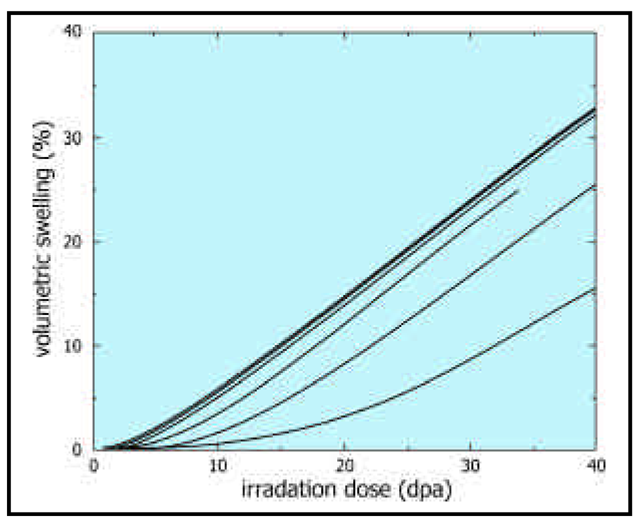



Figure 2. The graph plots volumetric swelling, $D$V/V in percent, versus total irradiation dose (equivalently, the irradiation time, in Msec) for a pure, type-316 stainless steel. The system parameters are exactly as in Figure 1, except that here, the dislocation bias factors are allowed to vary with the dislocation density. Again, the lowest temperature curves display the longest incubation delays.

dependent, and the final swelling rates are strongly bias-dependent.

There is a striking difference in the predicted number density of voids between the constant and the variable dislocation-bias simulations, The predicted terminal void population is several times smaller with the variable dislocation bias than in the constant dislocation-bias case (Figure 1).

In summary, the present microstructural code for void nucleation and concurrent dislocation network evolution is generating swelling-fluence-temperature correlations which closely resemble that obtained by Garner from recent data sets on EBR-II irradiated 304 stainless steels. At the same time, the new correlations are dramatically different than the empirical swelling design equations still in use for LWR and breeder reactors. In particular, it was found that irradiation temperature does not have a large effect on the steady-state swelling rate, but does have a large influence on the incubation dose.

## Planned Activities

During the initial stage of dislocation evolution in solution-annealed stainless steels, a high density of small dislocation loops are present. While these loops merge with the network dislocations at higher doses, they are expected to influence the void nucleation, and hence the incubation dose for void swelling.

During FY02, appropriate models will be developed for loop evolution and will be incorporated into the microstructural evolution code. Additional algorithm development will be undertaken to improve the numerical stability of the code and thereby improve the efficiency.

Collision cascades lead to atomic mixing in materials with small precipitates. An analogous effect should lead to the destruction of small void embryos. This should result in an increase of the incubation dose for void swelling with increasing neutron energy. To quantify this potential effect of the neutron energy spectrum on incubation, collision cascade simulations will be carried out near small clusters of vacancies with and without helium.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Development and Validation of Temperature Dependent Thermal Neutron Scattering Laws for Applications and Safety Implications in Generation IV Nuclear Reactor Designs

Primary Investigator: Ayman I. Hawari, North Carolina State University

Collaborators: Oak Ridge National Laboratory Instituto Balseiro, Argentina

Project Number: 01-140

Project Start Date: September 2001

Project End Date: September 2004

## Research Objectives

The overall objectives of this work are: to critically review the currently used thermal neutron scattering laws for various moderators and fuel cells as a function of temperature, to use the review as a guide in examining and updating the various computational approaches in establishing the scattering law, to understand the implications of the results obtained on the ability to accurately define the operating and safety characteristics (e.g. the moderator temperature coefficient) of a given reactor design -- that is, to know not only the reactivity coefficients but also their errors, sensitivity coefficients and covariance matrices, and finally to test and benchmark the developed models within the framework of a neutron slowing down experiment. In particular, the studies will concentrate on investigating the latest ENDF/B thermal neutron cross sections for reactor grade graphite, beryllium, beryllium oxide, zirconium hydride, high purity light water, heavy water and polyethylene at temperatures greater than or equal to room temperature. These materials are neutron moderators that will be used in the development of Generation IV nuclear power reactors and in many applications in the nuclear science and engineering field. Of major importance is graphite, which is the moderator in the modular pebble bed reactor (MPBR) that is being examined internationally as a possible Generation IV power reactor, as the subcritical reactor in accelerator driven concepts, and as the incinerator of radioactive waste and weapons' plutonium. Furthermore, a newly developed highly conductive form of graphite, known as graphite foam, is currently under study as a reactor material.

## Research Progress

During the first year, the effort of this project has concentrated on establishing the methodology and the tools that are required for evaluating the thermal neutron scattering laws for the materials of interest. Due to the recent interest in high temperature gas reactor (HTGR) concepts, including the pebble bed reactor, the research focused on evaluating the scattering kernel for graphite. This included updating it by implementing previously unused phonon frequency distributions, exploring the use of "ab initio" methods as a general approach for generating the phonon frequency distributions for crystalline moderators, identifying a set of experimental benchmarks to test the new kernel, and designing the temperature dependent neutron slowing-down-time experiment that will be used to test and validate the standard (i.e., ENDF/B-VI based) and the new thermal neutron scattering kernels for graphite.

As a starting point, the LEAPR methodology of the NJOY99 code system was used to reproduce the graphite scattering kernel using the phonon distribution function established by Young and Koppel in 1965, which is the basis of the ENDF/B-VI data. Once that was achieved successfully, the Young-Koppel data were replaced by a phonon frequency distribution function that was established by Nicklow, Wakabayashi, and Smith at Oak Ridge National Laboratory (ORNL) in 1972. While the general features of the ORNL phonon distribution are similar to the Young-Koppel phonon distribution, the detailed features are quite different. It was found that the resulting "scattering law" S(a,ß) curves using the ORNL phonon distribution for moderator temperatures 296°K, and 1,200°K were shifted to somewhat higher values compared to the Young-Koppel results. The inelastic cross sections for the two different phonon distributions were determined using the THERMR module of NJOY99. The figure below shows these results for graphite moderator temperatures of 296°K and 1,200°K.

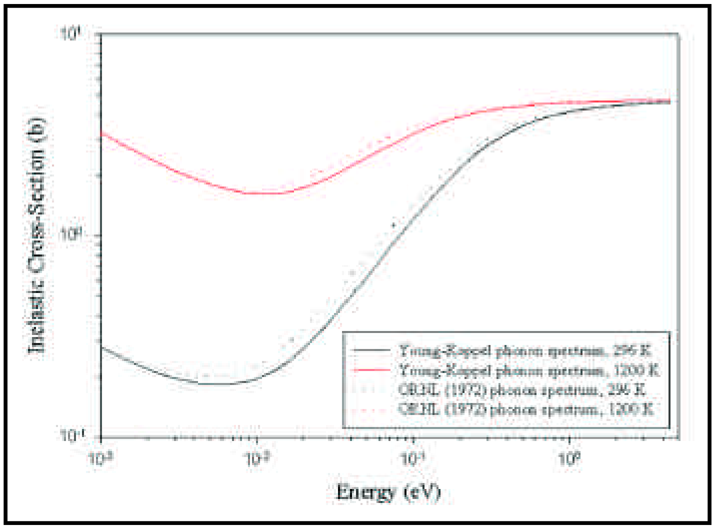



Figure 1. The graph illustrates an inelastic cross-section for graphite at 296°K and 1,200°K

Subsequently, the above data were formatted to be used in the MCNP code using the ACER module of NJOY99 and are currently being tested in computational HTGR benchmarks for sensitivity calculations of the values of keff and the total capture and reaction rates of actinides as a function of fuel and graphite temperatures.

Furthermore, we have initiated the use of the ab intio code VASP coupled to the PHONON code to establish the phonon spectra of solid moderators based on first principal quantum mechanical calculations. The work during this phase concentrated on optimizing our graphite models and subsequently applying the developed methodology to Be, BeO and ZrH.

In addition, we began a series of Monte Carlo simulations to design an experiment to benchmark the thermal neutron scattering kernel of graphite as a function of temperature. The simulations were performed using the MCNP code and its standard libraries. The simulated temperatures ranged from 300°K to 1,200°K. The model was based on introducing a neutron pulse at the surface of a 75 x 75 x 75 cm graphite pile and monitoring the time dependent reaction rate in a detector placed outside the pile at distances ranging from 0.25 m to 2 m from the surface opposite to the source. The neutron source was assumed to be the Oak Ridge Electron Linear Accelerator (ORELA). To create a realistic model, we assumed that the graphite pile would be located at the end of one of ORELA's beam tubes. Therefore, to ensure maximum source intensity, we chose the shortest beam tube, which is 10 m long. In addition, we assumed a 20-nanosecond pulse width and a 1000 Hz frequency. Under such conditions, the source strength at the surface of the pile is expected to be approximately 108 n/s. Furthermore, upon analyzing the ORELA energy spectrum we found that despite its spread, it is, in effect, a fast neutron spectrum peaking around 600 keV in ln(E). This only becomes apparent if the spectrum is expressed as (ln(E)) vs. E as opposed to the usual (E) vs. E. Based on this model, the time dependent reaction rate in a Pu-239 detector was calculated. The figure below shows the time spectra at a distance of 0.25 meters from the pile surface and for temperatures of 300, 800, and 1,200oK. The general features of the time spectra are dictated by the time dependent neutron energy spectrum that is leaking from the pile and the energy dependent fission cross-section of Pu-239. The close coupling of the leakage energy spectrum to the time after the neutron pulse can be predicted based on the principals of time dependent neutron slowing down theory. For both temperatures, the time spectra are nearly identical at early times (i.e., high energies). However, once thermal effects become important (around 10-4 seconds), the spectra begin to deviate due to the temperature dependence of the thermal neutron scattering kernel.

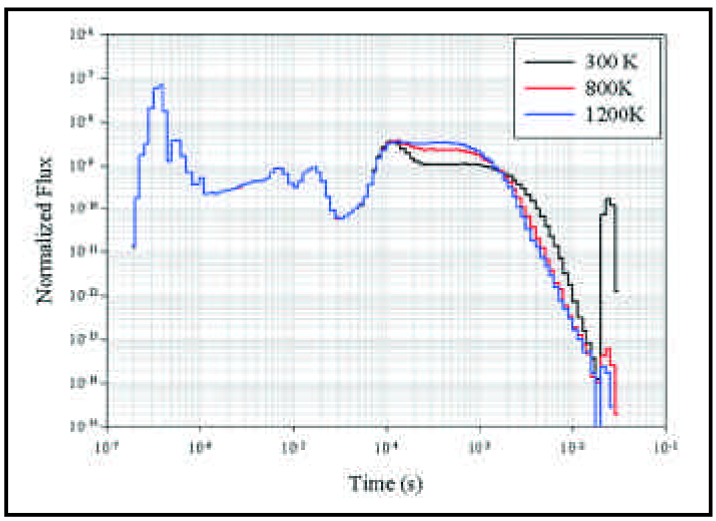



Figure 2. The plot is of the calculated time dependent reaction rates for a Pu-239 detector

## Planned Activities

During the upcoming phases of this project, work will continue on exploring new methods for generating the scattering kernels for the proposed materials and comparing them to the standard kernels. This will include ab initio methods and the synthetic kernel approach. In addition, computational benchmarks will be used to examine the validity of the new data and methods relative to current knowledge. During phase 2, this work will extend to understanding the implications of the results obtained on the ability to accurately define the operating and safety characteristics of a given reactor design. Work will also continue on the design of the graphite benchmark experiment. Preliminary experiments, at ORELA, are expected to begin during phase 2 of this project. The final experiments are planned to take place during phase 3 of the project.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Oxidation of Zircaloy Fuel Cladding in Water-Cooled Nuclear Reactors

Primary Investigator: Digby MacDonald, Pennsylvania State University

Project Number: 02-042

Project Start Date: September 2002

Project End Date: September 2005

With the development of higher burn-up fuels for nuclear power reactors, much greater demands are being placed on the performance of the Zircaloy fuel sheaths. The principal threat to the integrity of the sheath is oxidation/corrosion and hydriding, leading to somewhat uniform thinning, and in some instances to localized corrosion in the form of nodular attack and/or hydriding. Failure leads to the release of fission products into the coolant, which in turn contributes to the man-REM costs of operating the system. Extensive fuel failures may require shutdown, which results in the unit being unavailable for normal operation. Thus, strong operational and economic reasons exist for enhancing fuel reliability. The principal goal of the proposed work is to develop sophisticated physico-electrochemical models for the corrosion of Zircaloy fuel sheaths that can be used by reactor operators to actively manage the accumulation of damage and thereby minimize the risk of fuel cladding failure in operating reactors.

While the kinetics and mechanisms of the oxidation of zirconium and zirconium alloys have been extensively studied, little effort has been made to develop deterministic (as opposed to empirical and semi-empirical) models that can be used to predict fuel sheath performance and reliability at very high burn-ups in operating reactors. Those attempts at developing algorithms have employed semi-empirical, parabolic, or cubic models (e.g., $L2 = kt + C$, where L is the oxide thickness, t is time, k is the parabolic rate constant, and C is a constant) to extrapolate oxide thickness data to longer times, but the validity of these models is highly uncertain. Furthermore, the models generally ignore the cathodic reaction(s) that occur on the fuel sheath surface and none attempt to model the actual electrochemical conditions that exist within porous deposits at the fuel sheath/coolant (fs/c) interface. Other problems include the simplicity of the diffusion model that predicts parabolic growth, particularly when viewed in light of more modern models for the growth of anodic oxide films; the inability of the models to predict the transition that occurs in growth kinetics from parabolic (or cubic) to linear at a more-or-less specific oxide film thickness in terms of fundamental, atomic scale processes; the exclusion of effects due to second phase particle (SPPs) on the cathodic processes that occur within the outer layer; the lack of an atomic scale model for the formation of hydrides; and the inability of the present models to describe the influence of solution-phase species, such as Li+, on the oxide film growth kinetics. On reflection, it is apparent that the most glaring deficiency in the current theories and models is the lack of a sound electrochemical basis for the corrosion process under free corrosion conditions. By focusing only upon the oxidation of zirconium, these models in effect attempt to treat only half of the problem, in that the cathodic processes are ignored. Because the cathodic processes must be included in order to satisfy the conservation of charge, the existing models are "non-physical" and hence cannot constitute a deterministic basis for describing the oxidation phenomenon. Furthermore, some evidence exists to suggest that the cathodic reactions, which must occur at the same rate as the zirconium oxidation reaction, may actually control the overall rate, with the apparent dependence of the rate on oxide thickness reflecting the rate of electron transfer across the film.

The issue with respect to the underlying mechanisms of oxidation and hydriding is important and timely, because of the considerable advances that have been made on these subjects over the past several years. For example, work by the authors over the past twenty years, under DOE/BES sponsorship, has developed the Point Defect Model (PDM) for the growth and breakdown of anodic passive films that form on metal surfaces. Recent work has shown that the PDM provides a much better description of oxide film growth than does the classical "high field" model [HFM], and indeed one attempt has already been made to apply the PDM to the oxidation of Zircalloy-4. Furthermore, the formation of hydride is readily described by the PDM and a strong possibility exists that a unified, predictive model may be developed for oxidation and hydriding.

As noted above, electrochemical effects are almost totally ignored in the current models, but as with all corrosion processes they are actually dominant. Thus, the radiolysis of the coolant produces a number of electroactive species, including $H_2$, $O_2$, and $H_2O_2$, which react at the cladding/environment interface to consume the charge that is produced by the oxidation of the Zircaloy. The conservation of charge requires that the sum of the anodic partial currents due to Zr and $H_2$ oxidation be equal to that for the reduction of oxygen and hydrogen peroxide and any other reducible species in the system. The potential at which this condition is satisfied defines the corrosion potential (ECP), which is known to have a major impact on the corrosion of materials in reactor coolant circuits. Over the past decade, the author and his colleagues have developed sophisticated models for the radiolysis of water and the electrochemistry of the coolant circuits in boiling water reactors (BWRs) and more recently in pressurized water reactors (PWRs). Similar models will be developed in the proposed work to accurately describe the electrochemical conditions that exist at the cladding/coolant interface. The objectives of this program are to develop fundamentally new mechanisms for Zr oxidation and hydriding in reactor primary coolant environments that address issues arising from the specific chemistry employed (BWR vs. PWR) as well as from reactor-specific issues related to the mode of operation. The mechanisms will include the important phenomenon of boiling within porous CRUD deposits that exist on the fuel surface. This work is expected to yield new technologies for predicting the rate of growth of $ZrO_2$ on Zircaloy under high burn-up conditions that can be used to the great benefit of the U.S. nuclear power industry. The technologies will be based on recent advances that this team has made in discerning the mechanism(s) of oxidation and hydriding of metals and alloys, as well as on advanced models that will be developed to describe the electrochemistry of reactor coolants at the fuel cladding/coolant interface, in terms of the bulk coolant chemistry, interfacial boiling, and the operating conditions in the reactor. Thus, the goal is to produce a predictive model that relates fuel sheath performance to coolant chemistry and reactor operating history, so that an operator can devise the most cost-effective operating strategies that minimize the risk of fuel failure. The technology will be produced in the form of an algorithm that is readily incorporated into power plant computers and hence can be used as both a risk assessment tool and as an operating planning guide.

The proposed research will represent a major departure from work being carried out elsewhere, by undertaking the following activities:

- Incorporating the PDM in place of the diffusion models to describe oxide and hydride growth
- Incorporating the cathodic reactions that occur at the fuel cladding/coolant interface including the role of intermetallic precipitates in the film as catalytic sites for these reactions
- Incorporating an advanced coolant radiolysis model for estimating the concentrations of electroactive species ($O_2$, $H_2O_2$, $H_2$, etc) at the cladding surface, as a function of the chemistry of the coolant (pH, [Li], [B], [$H_2$]) and the operating conditions of the reactor
- Including mechanisms (cation vacancy condensation) for passivity breakdown as a means of describing the onset of nodular attack
- Developing a model based upon the generation and annihilation of point defects (oxygen vacancies, cation vacancies, and zirconium interstitials) at the Zircaloy/zirconia and zirconia/solution interfaces to describe the generation of stress in the interphasial region
- Incorporating a model for the concentration of solutes into porous deposits (CRUD) on the fuel under boiling (BWRs) or nucleate boiling (PWRs) conditions, in order to more accurately describe the environment that is in contact with the Zircaloy surface
- Integrating the damage over the operating history of the reactor, including start-ups, shut downs, and variable power operation
- Exploring the electronic structure and measure kinetic parameters for $ZrO_2$ film growth on Zircaloy under accurately simulated reactor operating conditions; following film growth in situ by electrochemical impedance spectroscopy (EIS)/capacitance measurements, while determining the electronic structure by using Mott-Schottky analysis

- Measuring kinetic parameters (exchange current densities and transfer coefficients) for the reduction of oxygen and the oxidation of hydrogen on Zircaloy under prototypical reactor operating conditions to accurately model the cathodic processes that occur on the cladding surface

The output of this project will be a more comprehensive understanding of the oxidation and hydriding of Zircaloy fuel cladding in reactor coolant environments, with particular emphasis on linkage between the plant operating parameters and the damage incurred due to oxidation and hydriding. Additionally, the project will yield a set of models and codes that will be made available to the nuclear power industry for managing the accumulation of corrosion damage to reactor fuel cladding as a function of the coolant chemistry and reactor operating conditions and history.

Finally, the development of the models and codes outlined in this proposal could greatly aid in the development of Generation IV reactors, by exploring water chemistry, materials/environment compatibility, and fuel design options that would minimize corrosion (oxidation and hydriding) damage to fuel cladding under specified operating regimes.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Incorporation of Integral Fuel Burnable Absorbers Boron and Gadolinium into Zirconium-Alloy Fuel Clad Material

Primary Investigator: K. Sridharan, University of Wisconsin

Collaborators: Sandia National Laboratory; Westinghouse Savannah River Company

Project Number: 02-044

Project Start Date: September 2002

Project End Date: September 2004

Long-lived fuels require the use of higher enrichments of $^{235}U$ or other fissile materials. Such high levels of fissile material lead to excessive fuel activity at the beginning of life. To counteract this excessive activity, integral fuel burnable absorbers (IFBA) are added to some rods in the fuel assembly. The three commonly used IFBA materials are gadolinium oxide and erbium oxide, which are added to the $UO_2$ powder, and zirconium-diboride that is applied as a coating on the $UO_2$ pellets using plasma spraying or chemical vapor deposition techniques. These operations are performed as part of the fuel manufacturing process in the fuel plants. Due to the potential for cross-contamination with fuel that does not contain IFBA, these operations are performed in a facility that is physically separated from the main plant. These operations tend to be very costly because of their small volume, and can add from 20 to 30 percent to the manufacturing cost of the fuel. Other manufacturing issues that impact cost are maintenance of the correct levels of dosing and reduction in the fuel melting point due to additions of gadolinium and erbium oxide.

The goal of the proposed research is to develop an alternative approach that involves incorporation of boron or gadolinium into the fuel cladding material rather than as a coating or additive to the fuel pellets. This paradigm shift will allow for the introduction of the IFBA in a non-nuclear regulated environment and will obviate the necessity of additional handling and processing of the fuel pellets. This could represent significant cost savings and potentially lead to greater reproducibility and control of the burnable fuel in the early stages of the reactor operation.

To achieve this objective, state-of-the-art, ion-based, surface engineering techniques will be applied. This will be performed using the IBEST (Ion Beam Surface Treatment) process being developed at Sandia National Laboratories, which involves the delivery of high energy ion beam pulses onto the surface of a target material. These pulses melt the top few microns of the target material's surface. The melt zone then solidifies rapidly at rates in excess of $10^9$K/sec due to rapid heat extraction by the underlying substrate heat sink. This rapid solidification allows for surface alloying well in excess of the thermodynamically dictated solubility limits. This effect can be beneficially applied to the objectives of the proposed research for incorporating boron or gadolinium into the near-surface regions of Zircaloy-4 and Zirlo material used for fuel cladding. Several variants of this approach will be investigated with the goal of optimizing the process parameters to achieve the desired structure, composition, and compositional gradient in the near-surface regions of the Zircaloy-4 and Zirlo. Detailed materials characterization of the modified surface regions will be performed at the University of Wisconsin. The durability of the modified zirconium alloys against corrosion and oxidation will be tested in steam autoclaves at Westinghouse Science & Technology Department.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Neutron and Beta/Gamma Radiolysis of Supercritical Water

Primary Investigator: David M. Bartels, Argonne National Laboratory

Collaborators: University of Wisconsin

Project Number: 02-060

Project Start Date: September 2002

Project End Date: September 2005

Commercial nuclear reactors provide a source of heat, used to drive a "heat engine" (turbine) to create electricity. A fundamental principle of thermodynamics is that the higher the temperature at which any heat engine is operated, the greater its efficiency. Consequently, an obvious way to increase the operating efficiency and profitability of future nuclear power plants is to heat the water of the primary cooling loop to higher temperatures. Current pressurized water reactors (PWRs) run at roughly 300°C and 100 atmospheres pressure. Designs under consideration would operate at 450°C and 250 atmospheres, i.e., well beyond the critical point of water. This would improve the thermodynamic efficiency by about 30 percent. A major unanswered question is, what changes occur in the radiation-induced chemistry in water as the temperature and pressure are raised beyond the critical point, and what does this imply for the limiting corrosion processes in the materials of the primary cooling loop?

The cooling water of any water-cooled reactor undergoes radiolytic decomposition, induced by gamma, fast-electron, and neutron radiation in the reactor cores. Unless mitigating steps are taken, oxidizing species produced by the coolant radiolysis can promote intergranular stress-corrosion cracking and irradiation-assisted, stress-corrosion cracking of iron- and nickel-based alloys. These will alter corrosion rates of iron- and nickel-based alloys, and of zirconium alloys, in reactors. One commonly used remedial measure to limit corrosion by oxidizing species is to add hydrogen in a sufficient quantity to chemically reduce transient radiolytic primary oxidizing species (OH, $H_2O_2$, $HO_2/O_2$-), thereby stopping the formation of oxidizing products ($H_2O_2$ and $O_2$). It is still unclear whether this will be effective at the higher temperatures proposed for future reactors. While an earlier NERI project has investigated some of the most important radiation chemistry in supercritical water, there is no information at all on the effect of neutron radiolysis, which is the main source of the troublesome oxidizing species.

The collaboration proposed here is ideally suited to discover most of the fundamental information necessary for a predictive model of radiation-induced chemistry in a supercritical water reactor core. Electron pulse radiolysis coupled with transient absorption spectroscopy is the method of choice for measuring the kinetics of radiation-induced species, as well as product yields for fast electron and gamma radiation. The Argonne Chemistry Division's linac is capable of producing 20 MeV electron pulses of 30 picoseconds duration, and the principal investigators at Argonne have extensive experience in measuring transients on a nanosecond and sub-nanosecond timescale. The University of Wisconsin's Nuclear Reactor Facility is a very convenient source of neutron radiation that can be exploited for radiolysis experiments from room temperature to 500°C. The combined capabilities of these facilities will make it possible to create a quantitative model for water radiolysis in both current PWR systems and supercritical water-cooled plants in the future.

The subject of this proposal touches on several areas of research mentioned in the NERI call for proposals. At its heart, the work is fundamental chemical science, which can be applied to both current and future reactor problems, and other areas of endeavor such as supercritical water oxidation technology. The direct application to nuclear engineering research is the design of reactors with higher performance and efficiency. The work proposed here is a follow-up and extension of research in a previous NERI project (276) on the same subject.

# Nuclear Energy Research Initiative

## Innovative Approach to Establish Root Causes for Cracking in Aggressive Reactor Environments

Primary Investigator: S. M. Bruemmer, Pacific Northwest National Laboratory

Collaborators: GE Global Research Center; Electric Power Research Institute; Framatome ANP, Inc.

Project Number: 02-075

Project Start Date: September 2002

Project End Date: September 2005

The successful development of Generation IV nuclear power systems must address and mitigate several materials-degradation issues now strongly impacting existing light water reactors (LWRs) after very long periods of operation. In addition, the more aggressive radiation and environmental exposures envisioned for various advanced reactor concepts will require materials with improved high-temperature properties and resistance to cracking. Although previous fast reactor and fusion device programs have focused on the development of improved structural materials for their relevant conditions, no comparable effort has been directed toward the conditions unique to water-cooled fission reactors since the inception of nuclear-powered propulsion units for submarines. The paramount issues impacting both LWR economics and safety have been corrosion and stress-corrosion cracking in high-temperature water. These degradation processes have continued to limit performance as the industry has changed operating parameters and materials. Mechanistic understanding and non-traditional approaches are necessary to create durable corrosion-resistant alloys and establish the foundation for advanced reactor designs. Less down time and longer component lifetimes are the drivers motivating this research for both Generation III and IV nuclear power systems.

Proposed research will focus on the characterization of critical Fe- and Ni-base stainless alloys tested under well-controlled conditions where in-service complexities can be minimized. Quantitative assessment of crack-growth rates will be used to isolate effects of key variables, while high-resolution analytical transmission electron microscopy will provide mechanistic insights by interrogating crack-tip corrosion/oxidation reactions and crack-tip structures at near atomic dimensions. Reactions at buried interfaces, not accessible by conventional approaches, will be systematically interrogated for the first time. Novel mechanistic "fingerprinting" of crack-tip structures tied to thermodynamic and kinetic modeling of crack-tip processes will be used to isolate causes of environmental cracking. Comparisons will be made with results on failed components removed from LWR service (funded separately by industry collaborators).

The proposed research strategy capitalizes on unique national laboratory, industry, and university capabilities to generate basic materials and corrosion science results with immediate impact to next generation nuclear power systems. This proposed work will be integrated with existing NERI projects, with fundamental research funded by the DOE Office of Basic Energy Sciences, and with focused U.S. and international projects dealing with current LWR degradation issues. This leveraged approach will facilitate the revolutionary advances envisioned by NERI by creating a multi-faceted effort combining the basic and applied science necessary to drive mechanistic understanding and promote development of next generation materials that meet the performance goals of advanced reactors.

NUCLEAR ENERGY RESEARCH INITIATIVE

## Design of Radiation-Tolerant Structural Alloys for Generation IV Nuclear Energy Systems

Primary Investigator: Todd R. Allen, Argonne National Laboratory

Collaborators: Pacific Northwest National Laboratory; University of Michigan; Japan Nuclear Cycle Development Institute

Project Number: 02-110

Project Start Date: September 2002

Project End Date: September 2005

Under the Generation IV Reactor initiative, revolutionary improvements in nuclear energy system design are being pursued in the areas of sustainability, economics, and safety and reliability. To meet these goals, advanced nuclear energy systems demand materials that minimize resource use, minimize waste impact, improve proliferation resistance, extend component lifetime, and reduce uncertainty in component performance. Simultaneously, they will potentially operate in higher temperature environments, with greater radiation dose, and in unique corrosion environments compared to previous generations of nuclear energy systems. Additionally, the material choices must provide for construction and operating costs that allow the nuclear energy system to compete in the marketplace.

The irradiation performance of structural materials will likely be the limiting factor in successful nuclear energy system development. The limits of the structural and fuel-related materials determine the performance of new nuclear energy systems. Satisfactory performance in a nuclear energy system is unusually demanding. In addition to the best characteristics and performance of materials that have been achieved in other advanced high-temperature energy systems, nuclear energy systems require exceptional performance under high fluence irradiation. Based on experience, materials not tailored for irradiation performance generally experience profound changes in virtually all important engineering and physical properties because of fundamental changes in structure caused by radiation damage.

This project will develop and characterize the radiation performance of materials with improved radiation resistance. Material classes will be chosen that are expected to be critical in multiple Generation IV technologies. The material design strategies to be tested fall into three main categories: (1) alloying, by adding oversized elements to the matrix; (2) engineering grain boundaries; and (3) designing the microstructure/nanostructure, such as by adding matrix precipitates.

The materials to be examined include both austenitic and ferritic-martensitic steels, both classes of which are expected to be key structural materials in many Generation IV concepts. The irradiation program will consist of scoping studies using proton and heavy-ion irradiations of key alloys and tailored alloy condition, and examination of materials irradiated in BOR-60 to confirm charged particle results. Examinations will include microstructural characterization, and mechanical properties evaluation using hardness and shear punch and stress-corrosion cracking.

The teaming of Argonne National Laboratory, Pacific Northwest National Laboratory, and the University of Michigan joins together institutions with critical skills and demonstrated capability in evaluating irradiation performance, along with experience in water reactor and liquid metal fast reactor systems. This project builds on the successes of NERI projects being performed jointly at these three institutions over the last three years.

NUCLEAR ENERGY RESEARCH INITIATIVE

# Enhanced Control of PWR Primary Coolant Water Chemistry Using Selective Separation Systems for Recovery and Recycle of Enriched Boric Acid

Primary Investigator: Ken Czerwinski, Massachusetts Institute of Technology

Collaborators: Los Alamos National Laboratory; Florida Power and Light; Pacific Southern Electric and Gas Co.; (n,p) Energy, Inc.; University of California, Berkeley

Project Number: 02-146

Project Start Date: September 2002

Project End Date: September 2005

The economics of operating existing and advanced pressurized water reactors (PWRs) clearly identify that increasing nuclear fuel enrichment will produce more energy. To operate within the nuclear reactor safety requirements, the concentration of natural boric acid used as a flux chemical shim would have to be increased. Enriched boric acid (B-10) has a greater cross section than natural boric acid and is favored over natural boric acid. This occurs because of primary side-water, corrosion-cracking issues associated with the increased requirement for higher lithium hydroxide (Li-7) concentrations to maintain operational pH with an increased natural boric acid concentration. However, the cost of producing and using enriched isotopes such as B-10 and Li-7 requires a means to cost-effectively recover and reuse them.

Under the NERI category of fundamental chemistry, under fundamental science, work is proposed to develop and field test polymeric sequestering systems designed to efficiently and selectively recover enriched boric acid/lithium hydroxide from the primary coolant water of reactors. These advanced separation materials will reduce the cost of operating existing and advanced light water reactor systems by improving the chemical control of the primary reactor coolant. Contaminants present in the coolant system will be characterized regarding their potential for interfering with the selective recovery of B-10 and Li-7, and counter measures will be developed to mitigate their interference. Cost benefits will result from greater energy production per reactor unit, reduced operational radiation exposure, and protection from accelerated corrosion of critical core components.

# INDEX OF NERI PROJECTS

## FY 1999 Projects

## FY 2000 Projects



## FY 2001 Projects

## FY 2002 Projects